The Physics of Transmutation Systems: System Capabilities and Performances

Phillip J. Finck
Argonne National Laboratory
pfinck@anl.gov
FJ/OH, August 2002

Table of Contents

The Physics of Transmutation Systems: System Capabilities and Performances

Phillip J. Finck
Argonne National Laboratory
pfinck@anl.gov
FJ/OH, August 2002

Summary

The objectives of transmutation systems are reviewed. A classification of transmutation system is then discussed, and the technical capabilities of the most widespread systems are analyzed, first in a generic sense, later with a strong emphasis on practical examples. Comparative studies of a wide range of transmutation systems are summarized.

Introduction

This document is complementary to a document produced by Prof. Salvatores on "The Physics of Transmutation in Critical or Subcritical Reactors and the Impact on the Fuel Cycle". In that document, Salvatores describes the fundamental of transmutation, through basic physics properties and general parametric studies. In the present document we try to go one step further towards practical implementation (while recognizing that the practical issues such as technology development and demonstration, and economics, can only be mentioned in a very superficial manner).

Section 1 briefly overviews the possible objectives of transmutation systems, and links these different objectives to possible technological paths. It also describes the overall constraints which have to be considered when developing and implementing transmutation systems.

In section 2 we briefly overview the technological constraints which need to be accounted for when designing transmutation systems.

In section 3 we attempt to provide a simplified classification of transmutation systems in order to clarify later comparisons. It compares heterogeneous and homogeneous recycle strategies, and single and multi-tier systems.

Section 4 presents case analyses for assessing the transmutation performance of various individual systems, starting with LWR's (1. generic results; 2. multirecycle of plutonium; 3. an alternative: transmutation based on a Thorium fuel cycle), followed by Gas-Cooled Reactors (with an emphasis on the "deep burn" approach), and followed by Fast Reactors and Accelerator Driven systems (1. generic results; 2. homogeneous recycle of transuranics; 3. practical limit between Fast Reactors and Accelerator Driven Systems)

Section 5 summarizes recent results on integrated system performances. It focuses first on interface effects between the two elements of a dual tier system, and then summarizes the major lessons learned from recent global physics studies.

1: Why do we develop transmutation systems?

A priori, the answer to that question might appear to be very simple, in the line of "because we need to get rid of nuclear waste". In reality, this is a rather complex question, with ramification into strategic and political issues, and with strong consequences on technological paths.

The first most fundamental issue relates to the definition of waste. Certain countries such as the United States consider that the whole Spent Nuclear Fuel (SNF) is waste. These countries will tend to find bulk methods for reducing the potential impact of that waste (for example, direct geological disposal has long been favored in the US). Other countries, such as Japan and several European countries consider that certain elements, in particular Plutonium, are a resource and should be utilized to produce energy. These countries would tend to develop sophisticated transmutation systems, where the valuable elements are recycled into commercial reactors, whereas the other elements are disposed of in special burners.

The second issue relates to policy; whereas European countries and Japan routinely accept the separation (and subsequent recycle) of plutonium from SNF, the US has established a strong policy, related to the proliferation risk of separated plutonium, of not allowing the separation of pure plutonium streams. Consequently, the US has for many years developed separation and recycle technologies which keep all transuranic elements together, whereas European countries and Japan have strived to separate individual elements and burn them with specific technologies.

The third issue relates to “network effects”. Typically, a technology which has a strong installed base has a very strong competitive advantage when compared to a new and potentially better performing technology. This explains why a significant amount of effort in the past years has been devoted to recycle in LWR’s, even though physics studies indicate that transmutation is better accomplished in fast reactors.

The objectives of transmutation enterprises can also been radically different, and one can distinguish several approaches discussed in different countries:

- Repository volume reduction: in this approach, SNF would be separated into various individual elements or groups, each group has several elements, which would then be incorporated into specific waste forms or fuels for recycling. In particular, isotopes which produce high levels of heat in the short term such as Cs137 and Sr90 could be stored into engineered waste forms and would be allowed to decay until later disposal. In that case, partial transmutation (for example using MOX recycle in LWR’s) would be a method for disposing of the separated plutonium in an irradiated MOX assembly. Other transuranics would be disposed of in glass waste forms. Transmutation requirements in this case would be minimal and would be met with the technologies available today.

- Reduction of the proliferation risk: while the risk of proliferation is difficult to define in quantified terms, there is a potential risk that the plutonium (and possibly neptunium) in SNF might be diverted towards illicit applications. For that case, it is strongly desirable to eliminate a very large fraction (99% or more) of these elements. This leads to the development of recycle strategies either based on the deep burn of plutonium in specialized fuels, or to the multirecycle of plutonium in more standard fuel types. In both cases, complex physics strategies need to be implemented and several examples are discussed in this paper: the multirecycle of plutonium in LWR’s using the CORAIL concept, the deep burn of plutonium in gas cooled reactors, the use of advanced thorium based fuel cycles for plutonium burning, and the multirecycle of plutonium in fast reactors.

- Reduction of the long term risk to the public: two approaches are considered. The first is concerned with reducing the long term dose to the public at the boundaries of potential repositories, and must address both the source term and the transportation of radionuclides through the geosphere. The transmutation objectives are very dependant on the geological conditions of the waste disposal site. For example for the Yucca Mountain site in the US, the objectives might be to transmute the Neptunium chain (Np-237, Am-241, Pu-241), Tc-99, and I-129. The second approach is more global and is concerned with reducing the potential toxicity, regardless of possible transportation paths:

this is the concept of radiotoxicity reduction as described in Prof. Salvatores' paper. The development of technologies for meeting these objectives has been ongoing for the last two decades and is illustrated in this paper by several examples: the recycle of minor actinides in conventional reactors using specially designed fuels and transmutation targets, and the homogeneous recycle of all transuranics in fast reactors or accelerator driven systems.

2. Technological constraints

When designing transmutation systems, careful attention must be paid to the neutronics feasibility and to the practicality of the system. The demonstration of neutronics feasibility involves verifying that the neutron economy of the system at startup and in equilibrium can be maintained, and that general performances of the neutron multiplying systems are within the operability envelope: for example it is necessary to verify that the system can operate safely. While this is the first essential step, it by no means provides the demonstration that the system is attractive. Practicality studies are needed to demonstrate that the system can be implemented in a reasonable industrial context. These studies cover the whole fuel cycle, including reactors, fuel reprocessing and fabrication plants, and possibly enrichment plant; they attempt to examine in detail the technologies involved in the successive fuel cycle steps and measure the practical consequences of potential transmutation strategies on these technologies. It is customary to distinguish technological constraints in two categories:

- Constraints on existing technologies: it is frequently attractive to modify existing infrastructures for accommodating an enhanced transmutation mission. In that case it is necessary to verify that the new approach does not adversely affect the performance of existing components. A typical example is the conversion of once through LWR based fuel cycles to one recycle MOX schemes: while this conversion assumes the development of new separations and fabrication technologies and infrastructure, it also requires LWR's to operate with a new fuel type. Typically MOX fuels lead to a harder neutronics spectrum due to the high thermal cross sections of Plutonium; spectral hardening reduces the efficiency of control rods and soluble poisons, and special care must be taken to accommodate the safety consequences of this effect; also, power peaking effects occur in MOX fuel located adjacent to standard fuel, and specific design features must be implemented to reduce the peaks and operate the reactor at nominal conditions. Difficulties in the fuel cycle infrastructure also appear if MOX multirecycled is contemplated, and limits due to dose buildup and criticality safety must be accounted for. Schemes for multirecycling Plutonium using the existing infrastructure are described in Section 4.

- Constraints on innovative technologies: over the past decades, several new approaches to transmutation have been proposed. The best known example is the use of Accelerator Driven Systems using novel fertile free fuels. Preliminary studies of these novel systems systematically indicate overwhelming advantages, but often ignore difficulties in key parameters, such as system safety, fuel performance, and separations losses and availability of waste forms. The rule of thumb to use there is stated simply as: "In fuel cycle analyses, the devil is in the details".

3. Classification of transmutation systems

This section provides a brief glossary of some important concepts in transmutation technologies, but is by no means intended to be complete.

The first distinction is between **heterogeneous** and **homogeneous** recycle methodologies:

- Homogeneous approaches attempt to mix all fuel and transmutation materials into a single fuel form: typical examples are the MIX approach for LWR's developed in France

and the IFR approach for fast reactors developed in the US. In general homogeneous approaches have been shown to be practical for the recycle of Plutonium in thermal systems, and for the recycle of all Transuranics in fast systems. Two major difficulties usually limit the use of homogeneous approaches: first, reactor safety characteristics can be strongly degraded when certain isotopes are introduced in the fuel. Second, the irradiated fuels usually emit relatively large neutron and gamma doses, and require additional investments in remote handling technologies.

- Heterogeneous approaches physically separate the standard "driver" fuel which serves as a neutron source, from transmuting targets. The targets can be either in the same fuel assembly in segregated pins, or in distinct specifically designed assemblies. Typical examples are the current MOX approach used in Europe and in Japan, advanced MOX designs such as the CORAIL, and various target-based approaches for thermal and fast reactors. The major advantages of heterogeneous approaches is that they maximize the use of existing fuels and separations technologies and reduce the size of the investments needed for fabricating and treating specialized targets. Usually neutronics of heterogeneous systems are quite favorable: in particular in thermal reactors, heterogeneity can be used to facilitate neutronics designs (see the CORAIL example in section 4). The potential difficulty with heterogeneous systems is the need to develop very advanced technologies for target fabrication, processing, disposal, and handling.

The second distinction is between **single tier** and **multiple tier** systems:

- Single tier systems attempt to use a unique technology for transmuting all elements. Typical examples are ADS and fast reactor based transmutation approaches proposed in European, Japanese, and US programs. A strong advantage of these approaches is the integrated character of the resulting nuclear enterprise, and the limited number of new technologies which need to be developed. Strong disadvantages have also been mentioned: first, these schemes rely on relatively novel technologies which have not yet been tested in a commercial environment; second, they must handle rather unusual materials which might not be attractive to a commercial operator; third, a single tier system, used as the transmutation complement of a commercial tier, will make up about 40% of the total nuclear infrastructure. This is considered by some as too large of a reliance on technologies which might be difficult to commercialize.

- Multiple tier systems attempt to use commercial reactors to do a significant fraction of the transmutation task, and rely on specialized reactors to accomplish the final cleanup of the SNF. This is illustrated by the typical French and Japanese double strata approaches, where commercial reactors are used to burn down most of the Plutonium, whereas fast reactors or ADS's are used to transmute the remainder of the Plutonium and the Minor Actinides. The advantage of such systems is that it makes optimal use of the commercial reactors; in the most optimistic case only about 5% of the total nuclear infrastructure is made out of specialized non commercial machines. But it might also have non obvious but significant disadvantages. First, this approach necessitates the development of multirecycle strategies in LWR's (as a single MOX pass reduces the rate of accumulation of Plutonium by only 20 to 25%); while some recent studies (see section 4) do indicate that these strategies are feasible, there is of yet no evidence that they will be attractive to commercial operators. Second, there exist strong interface effects between the first and the second tier: as a higher fraction of the Plutonium is burned in Tier 1, the fuel fabrication, separations, and reactor operations in the second tier become more complex. While this issue is not yet fully understood and no optimum burn fraction in the first tier has yet been defined, some data are presented in Section 5.

The third distinction is between **burn-down** and **sustainable** modes of operations. The burn down mode corresponds to today's situation, where large quantities of Spent Nuclear Fuel have been accumulated and need to be disposed of; the transmutation system must accommodate

incoming streams of Transuranics and burn them down relatively rapidly. In a sustainable mode, the incoming stream would be depleted uranium, the system would be generating its own fissile material, and would burn down its own waste. In general these two approaches can be accommodated using the same sets of technologies, with adjustments made for example on the fuel compositions. The physics of the two systems are highly overlapping and will not be distinguished here.

The fourth and last distinction is between **deep burn** and **multirecycle** strategies. Most nuclear fuels have limited burnup capabilities, and only a fraction (say up to 30%) of the transuranics can be burned before the fuel reaches its damage limits and needs to be reprocessed and refabricated. In order to reach total destruction a large number of passes (using at each pass a decreasing volume and thus core fraction) is required. There are losses at each pass in the separations and fabrication steps, and these losses will end up going into waste forms for disposal. Nevertheless, certain fuel types, such as the TRISO fuel used in thermal gas cooled reactors, are specially engineered to reach extremely high burnups, and could possibly provide a high rate of Plutonium or Minor Actinide destruction with limited need for recycle and refabrication. Losses to the final waste form would then be equal to the non-transmuted fraction of the initial material. This approach is illustrated by the deep burn gas cooled approach described in section 4. The debate between these two radically different approaches is on-going; they pose very different problems to the core designer, where the deep burn designs must devise means to reach very high burnups with acceptable neutron economy, whereas the multirecycle designs must accommodate for the definition of equilibrium situations.

4. Assessment of the transmutation performance of several systems

In this section we describe the physics capabilities of various systems for recycling Transuranic elements. The first subsection (4.1) is devoted to LWR's and is comprised successively of a summary of generic studies performed by the French CEA, followed by two detailed examples: the capability for multirecycling Plutonium in PWR's (the CORAIL concept developed by CEA) and an alternate scheme using the Thorium fuel cycle as a support to burn Plutonium (this concept is being developed at Brookhaven National Laboratory). The second subsection is devoted to the "deep-burn" concept in gas cooled reactors (this concept is developed at General Atomics), and provides an alternative for burning Plutonium and possibly some Minor Actinides. The third section is devoted to fast reactors and is comprised of three parts: a summary of generic studies performed by the French CEA, followed by a description of a scheme based on the homogeneous recycle of all transuranics (this concept is developed by Argonne National Laboratory), and a final discussion of the practical limits between reactors and accelerator driven systems.

4.1 LWR based transmutation

4.1.1 Generic studies

This subtitle is only here for reference, as these studies have been well summarized in Prof. Salvatores's writeup.

4.1.2 The CORAIL approach for Plutonium and Transuranic Transmutation

4.1.2.1 Background

One focus area in nuclear technology research and development worldwide is the reduction of the amount of transuranic nuclides (TRU) in spent nuclear fuel stockpiles. For example, the United States DOE and the French CEA have entered into a collaborative agreement to study both thermal- and fast-spectrum reactor-based transmutation approaches. A 1998 estimate of the stockpile of spent nuclear fuel in the United States is 38,414 metric tons heavy metal (MTHM) [4.1], with an estimated TRU component of 392 MTTRU. It is the TRU component of the

disposed waste which contributes most significantly to the long-term radiotoxicity and dose. The nuclear power units currently operating commercially in the United States, and the majority of those operating around the world, are of a light water cooled and moderated (LWR) design. A conventional, low-enriched UO_2 assembly produces approximately 6.5 kg of TRU during its irradiation history in an LWR. Consequently, the current growth of the TRU stockpile coming from the U.S. commercial nuclear sector amounts to ~26 MTTRU/year. The reduction, or at least a slowing of the growth rate, of the TRU stockpile would benefit a nuclear waste repository by reducing the long-term radiotoxicity of the disposed waste; alternatively, the effective capacity of the repository could be increased.

The French nuclear program currently practices mono-recycling of the plutonium recovered from spent UO_2 assemblies in their pressurized water reactors (PWR). In this approach, mixed-oxide (MOX) fuel assemblies comprise roughly one-third of the core loading; it has been shown that a LWR core loading of 20-50% homogeneous MOX fuel assemblies can be tolerated without modifications to reactor systems (e.g., control rods) to maintain safety criteria [4.2]. The mono-recycling approach consumes ~25% of the Pu loaded in the MOX assemblies, providing a commensurate reduction of the long-term (i.e., at 100,000 years after disposal) waste radiotoxicity and slowing the growth rate of the TRU stockpile [4.3].

Multi-recycling would keep the Pu in the reactor fuel cycle and practically eliminate it from the disposed nuclear waste (limited to partitioning losses). However, because the Pu isotopic vector is “degraded” with irradiation (e.g., consumption of Pu-239 and buildup of Pu-240), multi-recycling requires an increase of the Pu loading to meet the operating cycle length requirements. After a few recycles, the Pu loading in a MOX assembly can exceed a known point (~12% Pu/HM) at which the coolant void coefficient turns positive [4.4].

Recently, France and the U.S. have investigated the deployment of the so-called CORAIL concept [4.5] in PWRs as a means of stabilizing (i.e., no net growth) the Pu, or possibly Pu plus minor actinides (MA), stockpile. The CORAIL assembly uses a heterogeneous fuel pin layout (UO_2 and MOX pins) in a design that is “retrofittable” to a typical PWR core. Even with multi-recycling, it has been found that the reactivity coefficients for a full-core loading of CORAIL assemblies are comparable to those of a reference UO_2-fueled core. This report summarizes evaluations of the CORAIL concept with Pu or Pu plus MA multi-recycling performed at the Argonne National Laboratory (ANL) [4.6, 4.7].

4.1.2.2 Characteristics of the CORAIL Assembly

As shown in Figure 4.1, one of the distinguishing features of the CORAIL assembly is its heterogeneous fuel pin configuration. Compared with a partial-core loading of MOX assemblies, the CORAIL assembly design exchanges heterogeneities *between* adjacent assemblies for heterogeneities *within* the assembly. This has certain advantages: notably, that the Pu-bearing pins reside in a more thermalized neutron spectrum, increasing the Pu consumption rate. Also, control rod worths are slightly less degraded than in a homogeneous MOX assembly.

However, a heterogeneous fuel pin configuration will yield sharp flux gradients within the assembly, introducing the disadvantage of localized power peaking factors that are higher than those observed in homogeneous (e.g., conventional UO_2) assembly designs. The CORAIL assembly design was optimized to mitigate this effect, resulting in a configuration of 180 UO_2 fuel pins in the assembly interior and 84 MOX fuel pins in the peripheral region [4.8]. Furthermore, burnable poisons can be utilized to reduce the power peaking. Even with a full-core loading of CORAIL assemblies, the fractional loading of MOX pins in a core fully loaded with CORAIL assemblies is 32%. Thus, as noted above, safety criteria are not expected to be adversely affected.

The 17x17 fuel pin lattice of the CORAIL assembly has 25 guide tube locations and an overall assembly size that is comparable to a standard 17x17 Westinghouse PWR assembly [4.9]. Thus,

the assembly can be loaded into current-generation PWRs, exploiting the fleet of commercial nuclear power units for reactor-based transmutation.

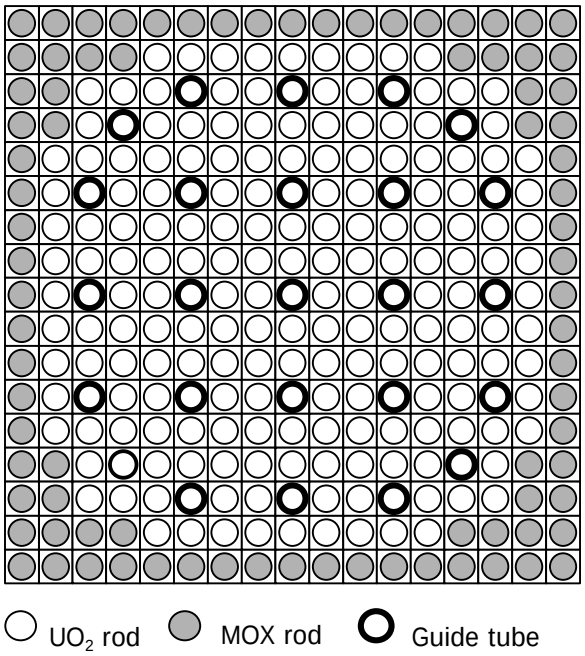

Figure 4.1. Fuel Pin Loading Pattern in the CORAIL Assembly.

4.1.2.3 Multi-Recycling in the CORAIL Concept

The CORAIL concept has been evaluated for multi-recycling of Pu (the so-called CORAIL-Pu assembly) and Pu plus MA (CORAIL-TRU). In the former case, the MA can be sent to a dedicated (fast-spectrum) transmutation system to complete the incineration of all TRU generated in the LWR fuel cycle. In the latter case, the entire transmutation mission is accomplished without the need for specialized transmuters, although practical issues may preclude complete incineration of the TRU in LWRs alone. In the following description of the multi-recycling approach, the generic label "TRU" may imply either Pu or Pu plus MA recycling.

Figure 4.2 presents a flow diagram for TRU multi-recycling in the CORAIL concept. For a given recycle, the MOX pins in the assembly are fabricated from TRU extracted from the discharge of the previous recycle (spent UO_2 and MOX pins); the fabrication of the UO_2 pins requires an external source of enriched uranium. A lead-time of two years is assumed from fuel separation/assembly fabrication to its loading into the reactor. After the assembly is discharged from the reactor, a five year post-irradiation cooling time is allowed before reprocessing the discharged fuel by aqueous and/or pyrochemical (i.e., dry) processing. It is assumed that during reprocessing, most of the TRU (99.9%) are recycled, while all fission products (FP) and 0.1% of the TRU are discharged as waste (in the case of Pu multi-recycling, the waste will include 0.1% of the Pu and 100% of the MA).

With each recycle, the TRU loading (and isotopic vector) in the MOX pins is effectively determined by the amount of TRU in the assembly discharged from the previous recycle (less separation losses). The TRU content will likely increase with each recycle until an equilibrium state is reached, in which the net TRU production in the UO_2 pins is balanced by the net TRU consumption in the MOX pins.
The uranium enrichment in the UO_2 pins must be determined prior to the assembly fabrication step, subject to a reactor cycle length requirement of 15 GWd/tHM. Constraints on the power peaking factor, UO_2 pin enrichment, and MOX pin TRU content were not rigorously applied, but

the evolution of these parameters with recycling was monitored; the latter two constraints are primarily related to enrichment plant limits. Two-dimensional, whole-core depletion analyses were simulated by assembly-level calculations with the WIMS8 code [4.10]. A 3% Δk leakage correction at the equivalent end of cycle (EOC) state was applied; assuming a 3-batch core reloading scheme and a linear reactivity model, the EOC assembly average burnup is 30 GWd/tHM (the assembly discharge burnup is 45 GWd/tHM). For full-core loadings of MOX assemblies, good agreement between whole-core and assembly-level calculations of discharge HM isotopic fractions has been observed.

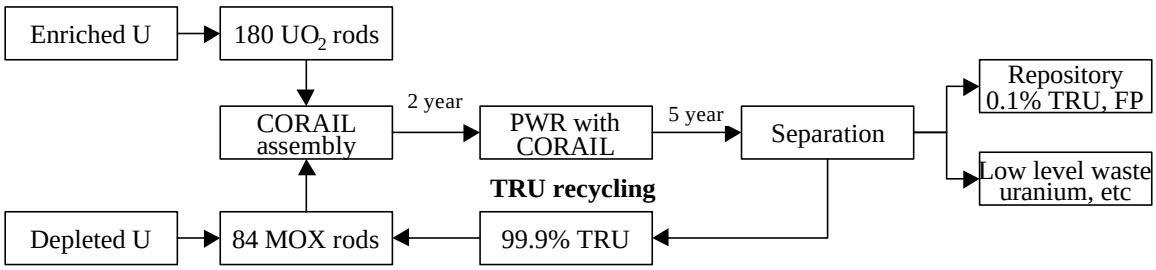

Figure 4.2. Flow Diagram for TRU Multi-Recycling with the CORAIL Concept.

Lastly, the CORAIL-fueled core is assumed to operate on an 18 month cycle. Thus, the assembly residence time (from charge to discharge) is 4.5 years. Including the fabrication lead-time and post-irradiation cooling interval, the total time for one recycle of the TRU is 11.5 years. Thus, even if TRU recycling is limited to just a few times (due, for example, to the complications associated with the accumulation of MA in the recycled material), the deployment of the CORAIL concept can provide a few-decade interval during which alternative transmutation technologies can mature.

4.1.2.4. Analysis of the CORAIL Concept

The attractiveness of the CORAIL concept for Pu or TRU stabilization has motivated interest in exploring the feasibility of its application in current and advanced PWRs. Mass flow and neutronics properties with recycling have been evaluated utilizing the computational algorithm outlined in the preceding section. The results for the 1st and 7th recycles are provided in Table 4.1; results for a conventional once-through UO_2 fuel cycle are provided as a reference.

The 1st recycle begins with reactor-grade Pu (63.4% fissile), as might be recovered from a spent UO_2 assembly. At the 7th recycle of the Pu, the MOX pin Pu loading is increased from 6.5% to 8.18%, while the fissile content of the vector is degraded to 46.8%; the assembly loading of fissile Pu decreases with recycling and is compensated by an increase of the U enrichment. The recycling of Pu plus MA begins in the same fashion. In this case, however, the MA are not discharged from the fuel cycle; instead, they accumulate in the recycled material. At the 7th recycle, the TRU content in the MOX pins is 12.74% (nearly doubled from that of the 1st recycle). The fissile TRU loading in the assembly is highest in this case, but the U enrichment must also be increased to compensate for parasitic capture in the MA.

The UO_2 assembly produces 6.0 kg Pu and 0.5 kg MA during its irradiation history, which would be added to the spent fuel stockpile. Deploying the CORAIL concept would reduce the Pu production rate by a factor of 4 to 7.5 (at the 7th recycle) for TRU and Pu recycling, respectively. Although the TRU stockpile is technically increasing with the CORAIL concept deployed (albeit, at a slower rate), practically the Pu is eliminated from the repository waste (limited to reprocessing losses) by keeping it in the fuel cycle. On the other hand, recycling the Pu produces 2.5 times more MA than the UO_2 assembly, raising the possibility that specialized MA transmuters [4.11] will be needed. If the MA are recycled, they will accumulate in the fuel cycle. Since high intrinsic

decay heat levels and neutron and γ sources make the MA difficult to handle, this accumulation will complicate fuel fabrication and handling; these issues will be addressed in a later section.

Local power peaking factors and reactivity coefficients were evaluated for comparison with a reference UO_2 assembly. The power peaking factor is notably higher in the CORAIL assembly, with the peak power typically occurring in the UO_2 pins. Burnable poisons could be utilized to flatten the assembly power shape; the impact of this on the assembly mass balances is yet to be evaluated. The reactivity coefficients for the CORAIL assembly compare well with those for the UO_2 assembly. The CORAIL soluble boron worth is less negative due to the harder spectrum in the TRU-bearing assembly, meaning that critical soluble boron concentrations may be higher in CORAIL-fueled cores. The coolant void worth is also less negative due to the higher Pu loading and displacement of U-238 from the lattice. An analysis of the impact of these coefficients on accident scenarios is certainly needed, but they do not appear to be problematic.

Table 4.1. Multi-Recycling Results in the CORAIL Assembly.

			CORAIL			**UO_2**
			1st Recycle	*7th Recycle*		
TRU Recycled			Pu	Pu	Pu plus MA	
Uranium Enrichment, %			4.15	4.57	5.04	4.0
TRU Content in MOX, %			6.50	8.18	12.74	-
BOC Power Peaking Factor			1.20	1.17	1.20	1.06
Initial TRU Vector ($^{w}/_{o}$)	Np-237				2.9	
	Pu-238		2.7	3.9	9.1	
	Pu-239		56.0	36.1	29.6	
	Pu-240		25.9	27.0	22.3	
	Pu-241		7.4	10.8	8.3	
	Pu-242		7.3	21.1	13.6	
	Am-241		0.7	1.1	5.6	
	Am-242m				0.1	
	Am-243				3.9	
	Cm-243				0.0	
	Cm-244				3.5	
	Cm-245				0.9	
	Fissile		63.4	46.8	38.9	
Mass Balance (kg/assembly)	Pu	Charge	11.0	13.8	18.0	0.0
		Discharge	11.7	14.6	19.5	6.0
		Net	**0.7**	**0.8**	**1.5**	**6.0**
	MA	Charge	0.1	0.2	3.7	0.0
		Discharge	1.1	1.4	3.6	0.5
		Net	**1.0**	**1.2**	**-0.1**	**0.5**
Reactivity Coefficients	Soluble boron worth (pcm/ppm)	Charge	-5.5	-5.3	-4.9	-6.7
		Discharge	-4.0	-4.2	-2.7	-9.4
	Coolant void worth (pcm/% void)	Charge	N/A	-243	-213	-259
		Discharge		-527	-402	-693

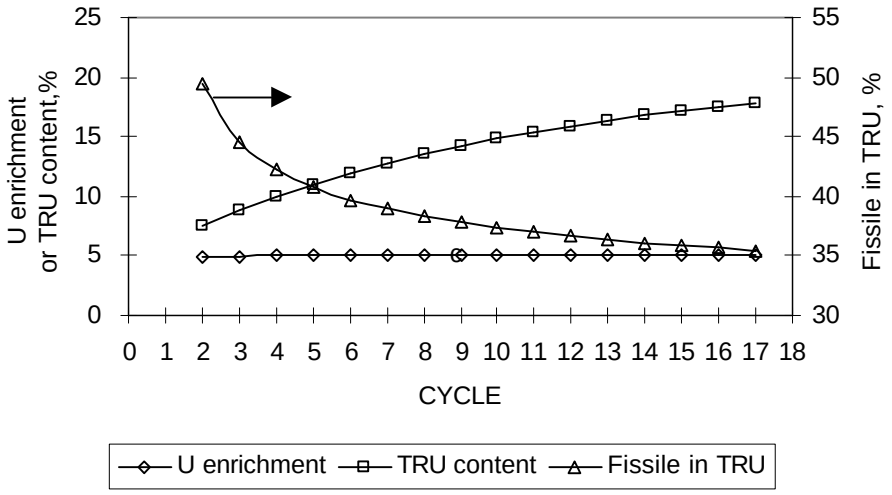

Figure 4.3. Repeated TRU Recycling in the CORAIL Assembly.

4.1.2.5 Impact of the CORAIL Concept on Repository Waste

One common measure of the effectiveness of transmutation approaches is the radiotoxicity of the disposed waste, normalized to that of the equivalent amount of uranium ore that must be mined to fabricate the fuel material. In the case of the CORAIL assembly, it was assumed that uranium ore is only needed to produce the enriched UO_2 pins; recycled TRU and depleted uranium are used to fabricate the MOX pins. In the CORAIL fuel cycle, the waste passed to the repository is limited to 0.1% of the recycled Pu and all fission products. Also, either 0.1% or all of the MA are disposed in the repository, depending on whether or not MA recycling is practiced; the present radiotoxicity analysis did not consider the impact of deploying MA transmuters if the MA are not recycled in the CORAIL fuel cycle. The discharged U will either be used as makeup feed or stored as low-level waste. On the other hand, all discharged heavy metal and fission products are passed to the repository in the reference UO_2 case, since spent fuel partitioning is not practiced in the once-through fuel cycle.

The normalized radiotoxicity, in terms of the cancer dose [4.12], was evaluated out to 10 million years after disposal for the waste from a reference UO_2 fuel cycle and the CORAIL fuel cycle with Pu or Pu plus MA recycling; the results of these evaluations are provided in Figure 4.4. The CORAIL evaluations are based on waste disposed between the 7^{th} and 8^{th} recycles.

With Pu recycling, higher concentrations of MA (particularly, Cm-244 and Am-241) in the waste elevate the radiotoxicity at ten years after disposal to slightly above (~10%) that for the UO_2 case. After that, the fission products (which contribute most significantly to the radiotoxicity) and Cm-244 decay rapidly to less radiotoxic daughters. By keeping the Pu out of the repository waste, the long-term radiotoxicity of the CORAIL waste is a factor of 2 to 5 lower. However, the disposal of Am-241 and other MA keeps the radiotoxicity above that of the natural uranium ore (normalized radiotoxicity > 1.0) out to 30,000 years after disposal.

If the Pu and MA are recycled together, the waste radiotoxicity is below that for UO_2 waste at all time points after disposal. Furthermore, the radiotoxicity of the disposed waste falls below that of the natural uranium ore after only 500 years because only a small fraction of all TRU (0.1%) is passed to the repository. It should be noted that if reprocessing losses are higher than the 0.1% assumed here, the length of time before the waste is less radiotoxic than the uranium ore can be significantly extended [4.13].

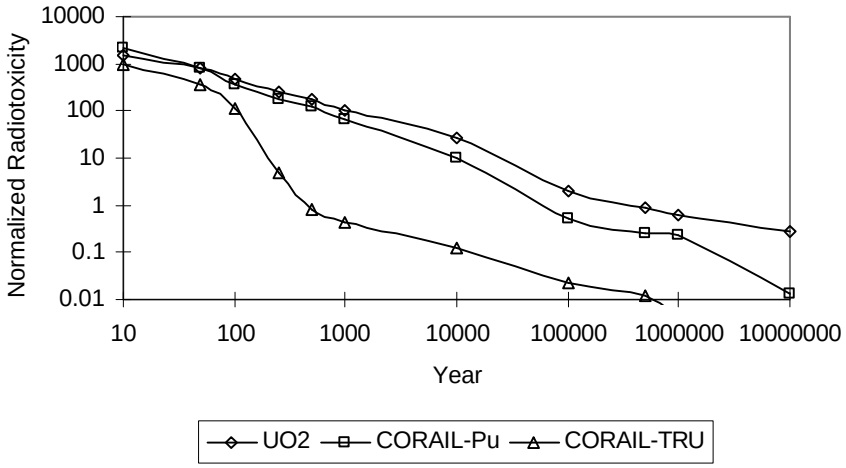

Figure 4.4. Normalized Radiotoxicity (Cancer Dose) of Disposed Waste

4.1.2.6 Fuel Handling Issues

The radiotoxicity of the nuclear waste stored in the repository environment is greatly reduced by recycling the Pu and MA in the CORAIL concept. However, the tradeoff for this benefit is an increase in the radioactive properties of the TRU-containing assemblies relative to a reference UO_2 assembly. This complicates fresh fuel handling (e.g. worker dose from neutron and gamma sources) and/or negatively impacts fuel separation processes (e.g. temperature increases due to higher decay heat loads may reduce process efficiencies).

As a preliminary effort to gain an understanding of the impact of TRU recycling on fuel handling and processing, three key parameters were identified and evaluated: total decay heat, γ energy emission rate, and neutron emission rate. Comparisons of these parameters for UO_2 and CORAIL assemblies at three stages in the reactor fuel cycle are provided in Table 6.1. The CORAIL evaluations are based on conditions in the 7^{th} recycle.

The UO_2 assemblies loaded into an LWR (i.e., at the "Charge" stage in Table 4.2) can be contact-handled by refueling operators; the decay heat, γ heat, and neutron source values are all very low since only long-lived uranium isotopes are present in the fresh fuel. The properties for the charged CORAIL assembly with Pu recycling are higher, but are not considered problematic; MOX assemblies fabricated with reactor-grade Pu, which contain roughly three times more Pu than the CORAIL assembly, are currently utilized in the French nuclear program.

However, recycling the MA along with the Pu increases the decay and γ heat values of the charged CORAIL assembly by nearly an order of magnitude, and the neutron source by a factor of 430. This is due to the higher assembly loadings of Cm-244 and Pu-238; Cm-244 has a relatively short half-life (18.1 years) and a small, but significant, branching ratio for spontaneous fission. The elevated decay heat would preclude contact handling, but the higher neutron source would require that additional measures (e.g., shielding and remote fuel handling) be implemented at the reactor to protect refueling operators.

After irradiation, the assemblies will be discharged from the reactor core and initially cooled in a spent fuel pool; the CORAIL assemblies would be cooled for five years before reprocessing. All assemblies have essentially the same total decay and γ heat loads at discharge (dominated by fission products), indicating that no special handling, beyond what is currently practiced for discharged UO_2 assemblies, is required for the discharged CORAIL assemblies. At the reprocessing stage (5 years cooled), the decay heat is highest when the MA are recycled, again due to the higher loading of Cm-244 and Pu-238. If aqueous processing is utilized, the higher

decay heat loads must be taken into account when optimizing the process to keep the temperatures of the process streams from being too high.

Table 4.2. Fuel Assembly Properties

		CORAIL		**UO_2**
TRU Recycled		Pu	Pu plus MA	N/A
Fabricated Assembly Mass (kg)				
HM		535	535	535
TRU		13.9	21.7	0
MA		0.2	3.7	0
Pu-238		0.5	2	0
Cm-244		0	0.8	0
Radioactive Properties				
Decay Heat (W)	Charge	**366**	**3,476**	0.0056
	Discharge	1.12E+06	1.10E+06	1.10E+06
	5 Years Cooled	2,160	5,400	1,440
γ Heat (W)	Charge	**0.2**	**1.8**	0.000044
	Discharge	3.11E+05	3.05E+05	3.12E+05
	5 Years Cooled	571	551	589
Neutron Source (n/s)	Charge	**2.00E+07**	**8.56E+09**	6.60E+03
	Discharge	4.64E+09	1.68E+10	6.71E+08
	5 Years Cooled	2.83E+09	1.09E+10	3.05E+08

The protection of workers at the reprocessing, fabrication, and reactor plants is most complicated by the high neutron emission rates which accompany MA recycling. Multi-recycling increases the Cm-244 loading, and the corresponding neutron emission rates (and decay heat values), in the TRU. Limiting the number of recycles will likely be necessary, but this will leave the TRU only partially consumed. Alternatively, lengthening the post-irradiation cooling interval would allow the Cm-244 ($t_{1/2}$=18.1 years) to decay to less-troublesome Pu-240 before reprocessing. Lastly, the Cm could be removed from the CORAIL fuel cycle and stored, and only the Np and Am recycled with the Pu. This would circumvent the buildup of Cm-244 in the CORAIL assembly, while still consuming the highly radiotoxic Am-241.

4.1.2.7 Conclusions

The CORAIL concept has been investigated as a means of stabilizing the TRU stockpile. The assembly design is retrofittable to a typical PWR core and has reactivity coefficients that are comparable to those of a conventional UO_2 assembly. Computational analyses have found that, from a physics perspective, Pu or Pu plus MA multi-recycling with the concept is feasible.

Deployment of the CORAIL concept in the current fleet of PWRs would reduce the rate of TRU-stockpile growth by a factor of 3-5. However, stabilization (i.e., zero net growth) of the stockpile will require many recycles to reach equilibrium and/or an optimized assembly design. Even so, the concept effectively keeps the TRU out of the disposed repository waste and provides an extended interval during which alternative transmutation technologies can mature. Furthermore, if the MA are recycled along with the Pu, the radiotoxicity of the disposed waste falls below that of its source uranium within 500 years, as opposed to >100,000 for conventional UO_2 waste.

However, multi-recycling of the TRU leads to a significant increase in the higher actinide content of the CORAIL assembly. This will complicate fuel handling (e.g. worker dose from neutron or γ sources) compared to standard UO_2 or MOX assemblies, and may be unfeasible in existing reactors. Thus, it is expected that *fuel cycle considerations* will constrain the extent of TRU recycle that can be practically achieved with the CORAIL concept.

4.1.3 Thorium-Based Fuels for Burning Plutonium

The thorium-^{233}U cycle offers several potential advantages relative to the conventional U-Pu cycle currently employed in commercial reactors including increased proliferation resistance (reduced Pu production and degradation of plutonium isotopics), improved fuel performance, and improved long-term waste characteristics. The proliferation potential of the bred ^{233}U is addressed by denaturing (adding uranium to the initial fuel composition to keep the ^{233}U concentration below the proliferation limit), and the high radiation field due to the high-energy gamma associated with the decay of U-232 and its daughters.

The potential of the thorium cycle to increase the net destruction of plutonium by reducing the secondary production of new plutonium, which is unavoidable in conventional UO_2-PuO_2 MOX, has also led to its consideration world-wide as a way to constrain the growth of plutonium. The promise of this option has resulted in several international studies focused on burning RGPu, most notably by the International Atomic Energy Agency (IAEA) [4.14], and the European Commission (EU) [4.15]. Similar improvements in the disposition of WGPu are expected.

In what follows, two conceptual PWR core designs are described whose objective is to reduce the potential for weapons-material proliferation by: 1) utilizing thorium as a fertile material, thereby reducing the production of reactor-grade plutonium, and 2) burning WG and RG plutonium, thereby reducing the stockpile of such material. As will be seen below, these approaches may be significantly superior to the approach presently being taken of mixing the plutonium with enriched uranium and burning mixed oxide (MOX) fuel in a portion of the fuel assemblies in the core.

Before discussing these designs the “ground rules” for evaluation are presented. Next there is a description of the two designs with most information given on the one with the greatest changes relative to existing PWR design. This is followed by a discussion of calculational results which show the features of the design and in particular the advantages for the objectives cited above.

In order to compare concepts one must first define the criteria that will be used. For the present study we use guidance from an IAEA study [4.14] which stated that the effectiveness for burning Pu could be quantified by considering:

- The amount of Pu which is burned per unit of produced electric energy. Maximization of this item optimizes the speed of the reduction of existing Pu stockpiles.

- The ratio of the amount of Pu which is burned during the lifetime of the fuel assemblies, to the amount of Pu which is residual in the unloaded fuel. Maximization of this item minimizes the quantity of Pu which either has to be finally disposed of, or has to be refabricated into fuel.

Maximization of the above quantities must be done in the context of a reactor that satisfies the normal operational and safety requirements for a nuclear power plant. Of particular concern are the safety parameters that are most affected by the fuel assembly and core design such as power peaking factors and reactivity feedback coefficients. In addition, it is necessary to consider the denaturing of the ^{233}U that is generated (to address non-proliferation objectives), and it may be desirable to assess the long-term toxicity of the discharged fuel relative to other approaches.

The two approaches that are considered as examples are: 1) the "Radkowsky Th-Fuel Pu Incinerator" (RTPI) [4.16], which is based on the RTF concept, and 2) the Korean Th-Pu PWR (KTPP) developed at the Korea Atomic Energy Research Institute [4.17]. The RTPI utilizes a seed-blanket unit (SBU) which is a one-for-one replacement for a conventional PWR fuel assembly, whereas the KTPP approach utilizes a homogeneous thorium fuel approach where each fuel rod in a PWR assembly contains ThO_2-PuO_2 fuel; this latter approach has also been evaluated by others [4.15]. These two approaches should be compared with the current approach in which MOX fuel is placed into a portion of the fuel assemblies.

Note that the two approaches considered herein are the most likely near-term options extant. There are other options for using Th and Pu with reactor concepts such as the pressurized heavy water reactor or the high temperature gas-cooled reactor, and there are other light water reactor (LWR) designs that use Th but not Pu as a way to limit the growth of RGPu. In the future, other LWR designs using both Th and Pu are expected to emerge.

Description of LWRs Using Thorium and Plutonium

The RTPI is a unique design optimized for the purpose of using both Th and WG/RG Pu whereas the KTPP is a more straightforward replacement of existing UO_2 with fuel containing ThO_2 and PuO_2. Both designs would replace standard 17x17 fuel assemblies with assemblies with the identical outer dimensions so that they would fit into existing cores without any modifications.

The RTPI fuel assembly design is based on the seed and blanket unit (SBU) described in Figure 4.5. Within the 17x17 fuel assembly the seed module is the central 11x11 subassembly and contains the Pu to be burned. The seed module is highly reactive neutronically and drives the remaining fuel located in the blanket surrounding the seed.

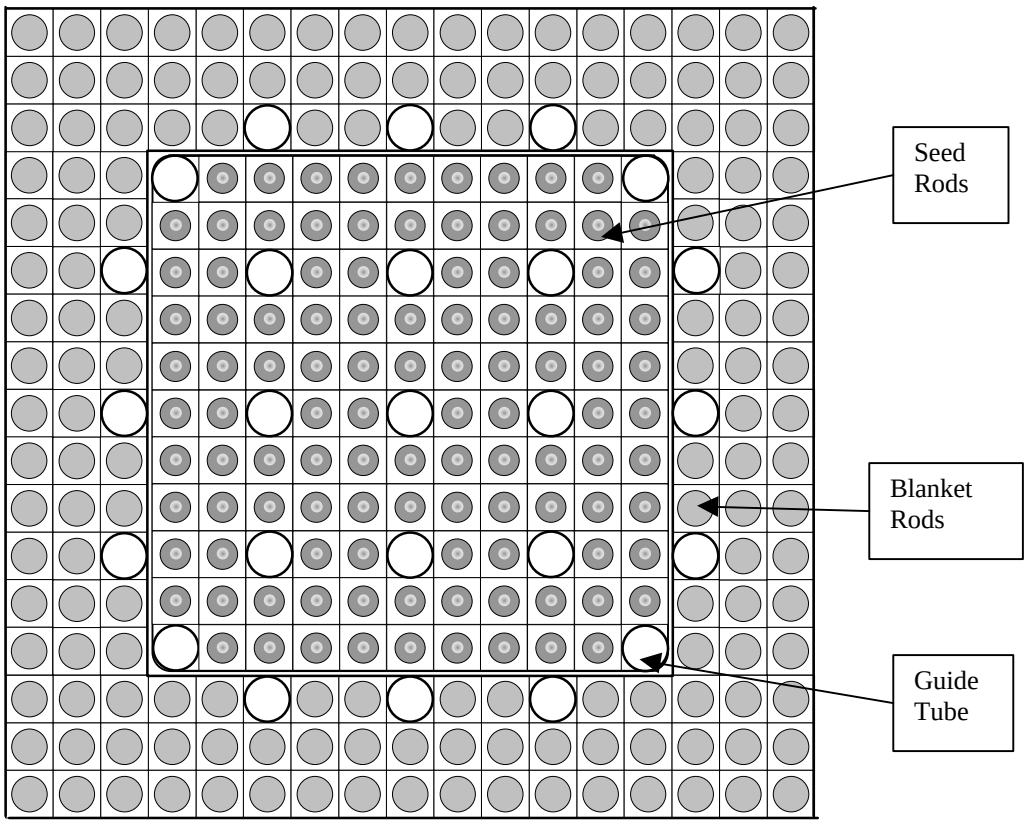

Figure 4.5. PWRT SBU Geometry

While the seed average power density (W/cc) is only slightly higher than that of a PWR, the specific power (MW/t) is considerably higher because of the relatively small amount of heavy metal in the seed. In three years of core residence, the seed fuel will acquire an average of 140,000 MWd/t. For oxide fuel this is well beyond current experience. Hence, metal fuel, specifically a Pu-Zr alloy, will be used as it is known that high burnups can be achieved with such fuel. Oxide fuel is also precluded due to the high power peaking and the need to have acceptable fuel temperatures, however, implementation of annular fuel pellet designs may alleviate this concern. The high heat conductivity of metal fuel, in conjunction with an annular design, with a Zr central region, reduces fuel temperatures to acceptable values. In the blanket the fuel dimensions and spacing are the same as in current PWRs.

The SBU geometry allows for separate fuel management for seed and blanket modules in the core. The seeds undergo the usual three-batch scheme, each seed residing for three years in the core, with one-third of all seed modules replaced annually. The blankets, however, reside for a full six years in the core for the RTPI. This longer residence time is designed primarily to allow the slow buildup of ^{233}U to take effect such that the blanket share in the core power is about 44%. Note that the average burnup at discharge for the blanket is only 63,000 MWd/t which allows for the use of oxide fuel.

The results for the key criteria for the two options described above as well as for a MOX core are given in Table 4.3 below. The results for the RTPI and KTPP are from the IAEA report [4.14] whereas the results for the MOX core are based on a study [4.18] reported on in Reference 4.16. In terms of the two criteria listed above, RTPI and KTPP burn approximately the same amount of Pu, however, the RTPI design has a higher ratio of Pu burned to discharged by approximately 33%. Clearly, both approaches are superior to using MOX fuel in a PWR as the amount of Pu burned and the ratio of burned to discharged Pu is relatively low for the MOX option.

Table 4.3. Mass Balance kg/GW_e-yr Using WG Pu

	RTPI	**KTPP**	**MOX**
Pu Charged	1095	1264	1000
Pu Discharged	361	507	740
Pu Burned	734	757	260
Burned/Discharged	2.0	1.5	0.35

These results are consistent with those from the EU study [4.15] which found that in general twice as much plutonium is consumed in thorium-based systems as with MOX.

The key core physics parameters for each of the core designs have been calculated and compared. In general, the temperature reactivity coefficients are comparable for all three designs and consistent with existing PWR designs. However, both the boron reactivity coefficient and the control rod worth are reduced in all three designs relative to existing PWR designs. This is especially true for the MOX design although still a serious consideration in the designs with Th.

For the homogeneous KTPP design no uranium is used whereas for the RTPI design uranium is used in the blanket with the thorium. In the RTPI this means that the ^{233}U will be denatured to an equivalent of <20% U-235 enrichment, the acceptance limit for non-proliferating fuel. Since the KTPP design does not consider denaturing the bred ^{233}U, a new proliferation concern results. This is the case despite the high-energy gamma from ^{232}U and its daughters which necessitates remote handling for any reprocessing. The question of denaturing will have to be addressed in the future. It should be noted that if denaturing is implemented to increase the proliferation

resistance associated with the bred ^{233}U, a more complicated ternary fuel of ThO_2-UO_2-PuO_2 would be required, and Pu-burning performance would be degraded.

For the two Th-Pu designs considered above, it is not clear from the limited documentation which might be superior. However, it should be noted that comparisons have been made of the SBU design relative to a homogeneous design for Th-U reactors and in those cases it was clear that the SBU design had significant advantages based on the nuclear design. [4.19]

4.2 Deep burn in gas cooled reactors

4.2.1 Background

The capability of the gas-cooled, graphite-moderated reactor to transmute or destroy the TRU waste discharged from Light Water Reactors (LWRs) are presented. The waste chosen for this analysis was assumed to be LWR recycled transuranic (TRU) discharged after about 40,000 MWD/MTHM burnup in the reactor, and stored for 10 years before processing. The discharge from this transmuter is being further transmuted by an accelerator driven system or fast reactor, or as a stand alone transmuter with its discharged waste being sent directly to the repository.

To maximize lifetime and burnup, a three segment core design was chosen, with one third of the core replaced at each refueling. Refueling intervals of 12 months, 18 months, and 24 months were evaluated, assuming 80% capacity factors. The 18 month cycle appears to be the best choice. About 78% of the input TRU waste, almost 99% of the fissile plutonium is destroyed. There is one reprocessing step after the initial separation of the TRU from the LWR discharge, thus minimizing reprocessing waste and worker exposure. This irradiation meets the fluence and burnup limits demonstrated in tests to date on coated particle fuels; although this performance capability needs to be confirmed by tests on the specific fuel compositions chosen for this analysis.

The 600-MWt, commercial Gas Turbine – Modular Helium Reactor (GT-MHR) design is used as the basis for this study, and the reactor design and layout for this system is described. A key requirement for the fuel cycle analysis is that this commercial GT-MHR reactor, designed for operation with a “standard” fuel cycle such as LEU, could utilize this LWR TRU waste as fuel and still meet all the normal operating, safety, and licensing conditions for the commercial plant. In other words the only change required to a GT-MHR would be to load the core with the LWR TRU fuel. This would minimize the development costs for this transmutation option. It also means that the transmuter fuel could be tested in small quantities in a commercial plant as it is being developed.

The fuel cycle studies and the transmuter analyses describe the physics of this transmuter and maximize LWR TRU transmutation and fissile plutonium destruction. For this purpose, the fuel cycle employs two different particles, a Driver Fuel (DF) consisting of the neptunium and plutonium from the TRU waste, and a Transmuter Fuel (TF) consisting of the rest of the minor actinides (americium and curium) from the TRU waste plus the transuranics discharged from the DF after its in-core irradiation. This fuel cycle uses the non-thermally fissile minor actinides as a burnable poison instead of wasting neutrons on a fertile material or other neutron poison. This maximizes both the Total TRU waste and fissile plutonium destruction, since most of the TRU (in the DF) is irradiated twice.

This transmuter can operate on other transmutation fuel cycles and different TRU compositions. In particular it can utilize just the plutonium (i.e., the DF), with a burnable poison such as erbium added for reactivity control. This option would allow for a longer development time for the TF, if necessary, while still allowing basic transmutation and energy production. By varying cycle time and fuel loading it should be possible for this transmuter to utilize a wide range of irradiated TRU waste as fuel. Thus it should be able to handle the wide range of LWR TRU waste from currently operating reactors

The analysis has not shown any feasibility issues. However, further studies are required to evaluate the selection of the most economical transmutation fuel cycle, and to determine the critical parameters for safety, performance, and operability for this cycle. Material flows, costs, proliferation risk, and residual toxicity would also be assessed.

4.2.2 System description

The transmuter [4-20] consists of a steel vessel housing, containing an annular transmutation region operating with a thermal neutron spectrum. This annular region contains the "fresh" TRU separated from light water reactor spent fuels. A general configuration of the system is shown in Figures 4.6 and 4.7. The TRU materials are contained in spherical TRI-material ISOtropic particle, TRISO-coated particles. These spherical particles consist of a TRU-oxide core, called kernel, surrounded by layers of graphite buffer to absorb gaseous fission products, pyrolitic graphite, silicon carbide to serve as a stable barrier and pressure vessel, and an outside layer of pyrolitic graphite. These particles are mixed with graphite powder and packed into cylindrical fuel compacts. The compacts are loaded into cylindrical channels within hexagonal graphite elements (blocks). Figure 4.8 shows the TRU particle details, fuel compact, and a fuel element. These blocks also have channels for helium coolant flow and channels for introducing burnable poison

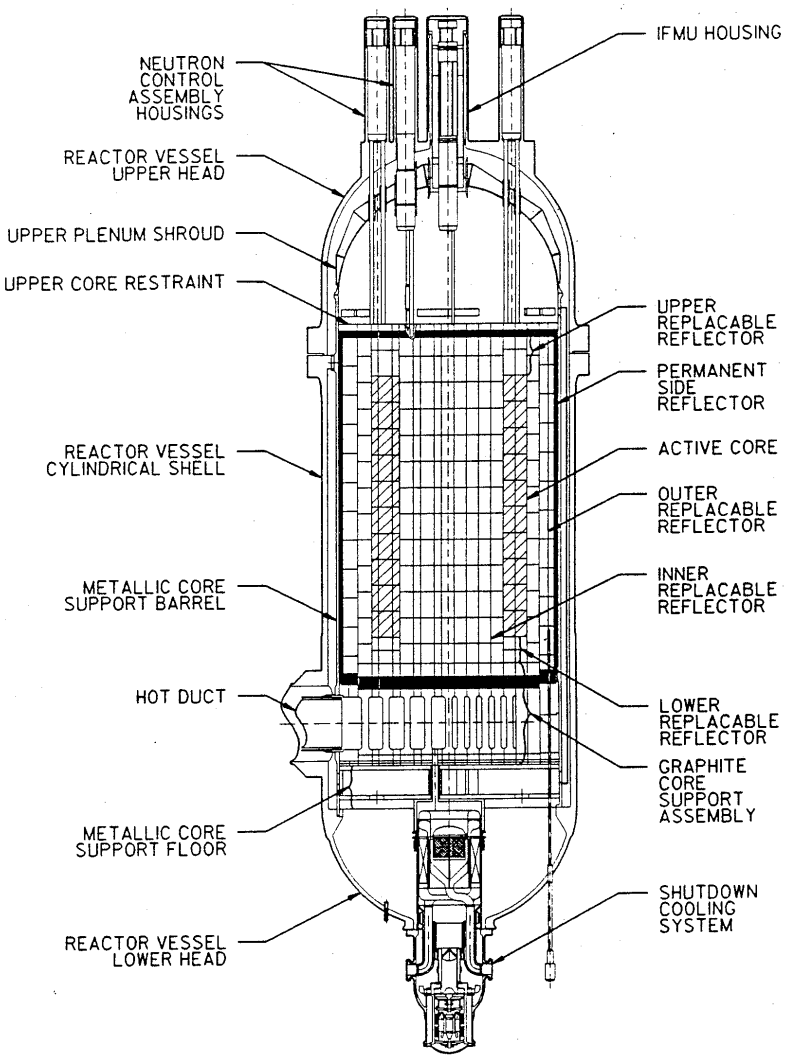

Figure 4.6. Reactor Elevation View

into the system. The block is 36 cm flat-to-flat, and contains 216 fuel/burnable poison channels, and 108 coolant channels. All the channels are arranged on a 1.88-cm triangular pitch. The fuel blocks are loaded into the fifth, the sixth, and the seventh radial rings of a hexagonal configuration. Graphite reflectors are arranged both inside and outside the fuel region.

The active core consists of the hexagonal graphite fuel blocks containing blind holes for fuel compacts and full-length holes for helium coolant flow. The fuel elements are stacked to form columns (10 fuel elements per column) that rest on support structures as shown in Figure 4.6. The active core consists of the three-row annulus as shown Figure 4.7. Eighteen of the core columns, and 30 of the outer reflector columns, contain channels for control rods. Eighteen columns in the core also contain channels for reserve shutdown control (RSC) material. It should be noted that the pattern of standard columns, control columns, and RSC columns has 1/3rd core symmetry.

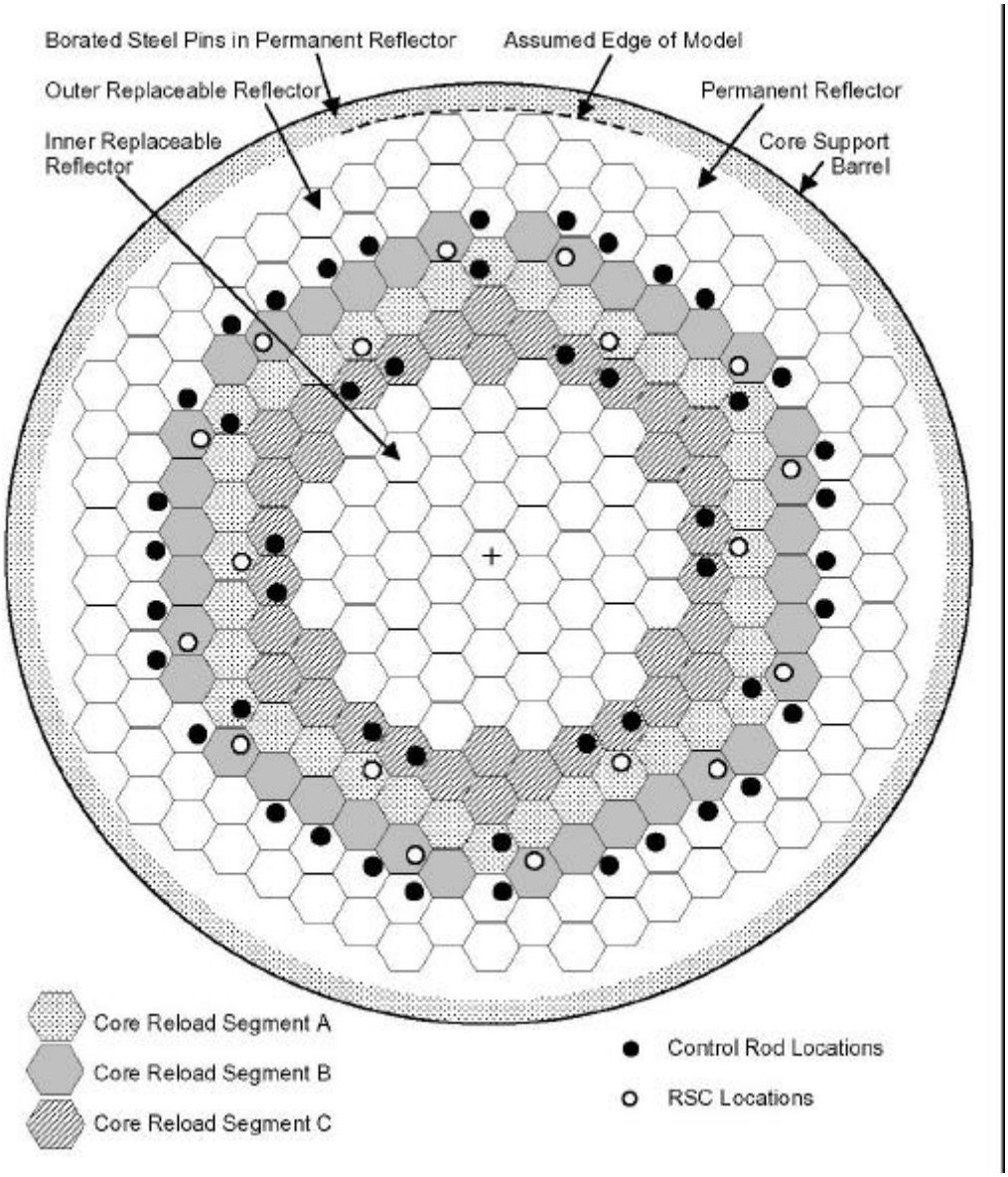

Figure 4.7. Core layout

There are two full rows of replaceable reflector columns radially outside of the core, and a permanent reflector outside of these two rows. The permanent reflector has a cylindrical outer boundary, with a region at the outside edge containing borated steel pins. This borated steel pin region is of uniform thickness around the reactor. The inside boundary of this borated steel pin region in the permanent reflector is assumed to just touch the replaceable reflector column furthest from the center of the core, as shown in Figure 4.7. There are replaceable reflector elements above and below the fueled core elements with borated steel pin regions at the very top and bottom of these axial reflectors. Basic core nuclear design parameters are summarized in Table 4.4.

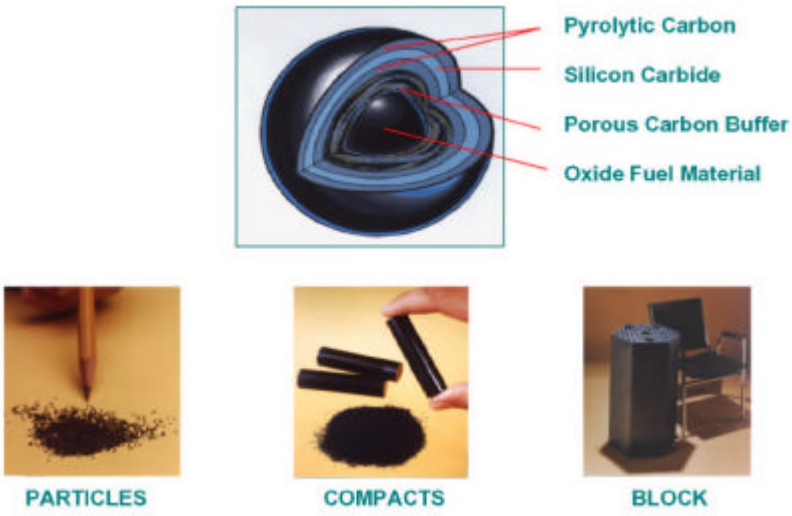

Figure 4.8. TRU particle, fuel compact, and block

The transmuter is cooled by helium with an outlet temperature of 850 °C. The heated helium is used in a direct-cycle gas-turbine-generator system. The high helium operating temperature and the utilization of the direct Brayton power conversion system allow electric generation with a high net thermal efficiency of ~50%.

4.2.3 Computational models and computer codes

The transmuter design includes several levels of heterogeneity effects that require proper treatment to obtain accurate performance predictions. The transmuter geometry consists of hexagonal prismatic blocks of graphite containing parallel vertical holes, arranged in a triangular pitch. These holes contain fuel or burnable poison (BP) compacts and some vacant holes for helium coolant flow paths. The fuel and the BP compacts consist of multi-layer ceramic-coated particles dispersed in a graphite matrix. Significant neutronics heterogeneities are created by these small particles. Block heterogeneity arises from the heterogeneous arrangement of fuel, BP, and coolant channels in the graphite block. Another geometrical heterogeneity is due to the annular configuration that employs inner and outer reflector zones.

Three independent computational paths have been implemented for the analyses:

1. An independent stochastic path based on MONK computer code [4.21] has been utilized. It is also used to check the predictions of the deterministic path and to provide a reference database. The MONK capabilities to explicitly model the geometry under consideration and to perform criticality and burnup analyses in an integrated manner were utilized.

Table 4.4. Core design parameters

Core Power, Mw(t)	600
Core columns	108
Elements per column	10
Number of control rods, inner reflector	0
Core	18
Outer reflector	30
Number of RSC in core	18
Standard elements in core	720
Control/RSC elements in core	360
Fuel segments	3
Element pitch, includes gaps between elements, cm (in)	36.10 (14.21)
Dimensions of hexagonal fuel element, cm (in.):	
Flat-to-flat, not including gaps between elements	36.0 (14.17)
Height	79.30 (31.22)
Active core height, cm (in.)	793.0 (312.2)
Control rod hole diameter, cm (in.)	10.16 (4.0)
RSC hole diameter, cm (in.)	10.16 (4.0)
Coolant holes per element, small/large:	
Standard element	6/102
Control/RSC element	7/88
Coolant hole diameter, cm (in.):	
Small	1.270 (0.50)
Large	1.5875 (0.625)
Fuel hole diameter, cm (in.)	1.270 (0.50)
Fuel hole length, under dowels, cm (in.)	75.2602 (29.63)
Not under dowels	78.1558 (30.77)
Compact diameter, cm (in.)	1.2446 (0.49)
Compact length, cm (in.)	4.9276 (1.94)
Compacts per fuel rod, under dowels/not under dowels	14/15
Pitch of fuel rod and coolant hole array	1.8796 (0.74)

An explicit detailed model for the block was developed using the MONK computer code. MONK has the capability to explicitly model the geometry under consideration and to perform criticality and burnup analyses in an integrated manner. The particles are modeled as a hexagonally close-packed lattice of spheres. The lattice forms a regular octahedron with a cylindrical boundary to represent the compact. MONK criticality calculations were performed with quasi-continuous energy and multigroup data sets. The quasi-continuous energy data sets are processed in a fine energy mesh (13193 or 8220 groups). The multigroup libraries are processed in a much coarser set (172 or 69 groups). The burnup analyses use the coarser data sets. The nuclear data libraries are based on JEF version 2.2 or ENDF/B-VI.

Three MONK models were developed for the heterogeneity analyses. These are:

- An explicit block model with explicit representation of the multi-layers of the TRU and the BP particles inside the compacts,
- A block model with homogenized particles inside the compacts, and
- A block model with homogenized compacts inside the block.

Table 4.4 and 4.5 give the data that are used to generate the MONK block models. Figure 4.9 shows the explicit MONK models for fuel particle, fuel block with burnable poison, an enlarged block section, and the horizontal cross section of the transmuter model.

The three-dimensional transmuter MONK model has explicit representation for the TRU and the burnable poison particles including all the geometrical details. The blocks are located in rings six to eight. Rings one to five and nine to eleven contain graphite reflector blocks. Axially, the whole length of the active core (793 cm), and additional lower and upper graphite reflector blocks are modeled. A vacuum boundary condition is used for all external surfaces. A cylindrical boundary is used for the radial reflector to match the actual configuration.

2. A deterministic path based on the DRAGON [4.22], DIF3D [4.23], and REBUS [4.24] computer codes has been utilized. It promises fast computer running times but it relies on a series of energetic and spatial homogenization steps, which might decrease the accuracy of the results. Thus, it needs to be carefully validated. Burnup-dependent, block-average microscopic cross-sections are obtained using the DRAGON lattice computer code and an ENDF/B-VI based 69-group library. The DRAGON computer code is selected because it models accurately the dispersion fuel in a graphite matrix and permits full-block calculations using the collision probability method. Resonance self-shielding and depletion calculations in the particles are possible because DRAGON allows explicit representations of the multi-layer TRU and BP particles, the matrix graphite, and the block graphite of the transmuter.

3. Another independent stochastic path based *on the* MONTEBURNS [4.25] computer code is utilized. It is a fully automated tool that links the Monte Carlo transport code MCNP [4.26] with the radioactive decay and the burnup code ORIGEN2 [4.27]. MONTEBURNS produces a large number of criticality and burn-up results based on various material feed/removal specifications, power(s), and time intervals. The program processes input from the user that specifies the system geometry, initial material compositions, feed/removal specifications, and other code-specific parameters. Various results from MCNP, ORIGEN2, and other calculations are then output successively as the code runs. The principle function of MONTEBURNS is to transfer cross-section and flux values from MCNP to ORIGEN2, and then transfer the resulting material compositions (after irradiation and/or decay) from ORIGEN2 back to MCNP in a repeated, cyclic fashion. The basic requirement of the code is that the user has a working MCNP input file and other input parameters; all interaction with ORIGEN2 and other calculations are then performed by MONTEBURNS. The nuclear data libraries are based on ENDF/B-VI release 2 processed with NJOY.

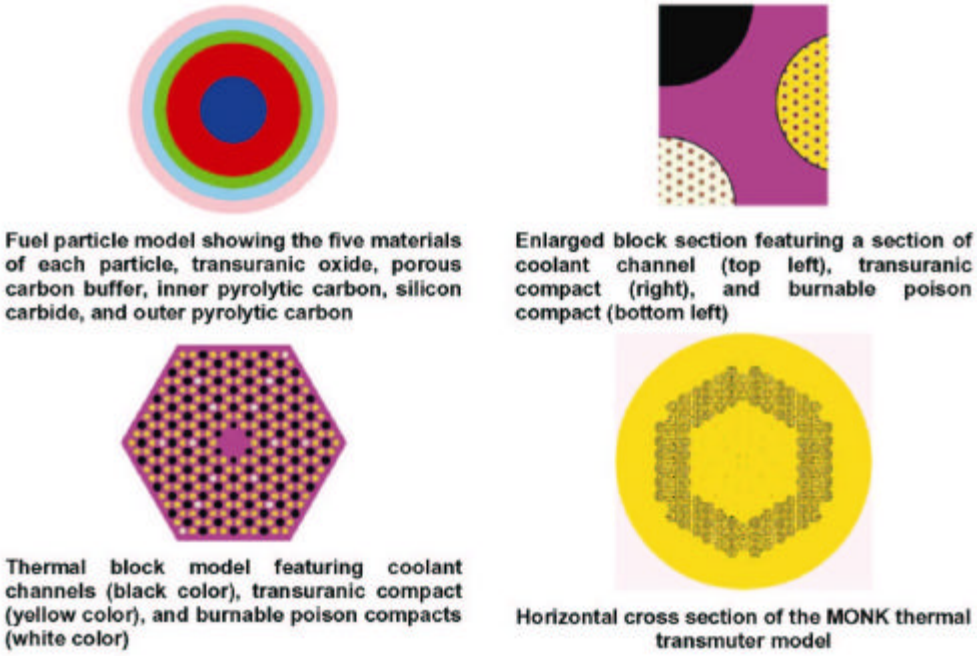

Figure 4.9. MONK geometrical models

Table 4.5. Particle parameters

Parameter	Value
TRU Kernel Properties	
Form, Density	$TRUO_{1.7}$, 10.2 g/cc
Diameter	200 µm
TRU Particle Coating Properties	
Buffer, Density	100 µm, 1.00 g/cc
Inner Dense PyC, Density	35 µm, 1.87 g/cc
SiC, Density	35 µm, 3.20 g/cc
Outer Dense PyC, Density	40 µm, 1.83 g/cc
Particle Diameter	620 µm
Erbium Kernel Properties	
Form, Density	Er_2O_3, 8.64 g/cc
Diameter	400 µm
Erbium Particle Coating Properties	
Buffer, Density	100 µm, 1.00 g/cc
Inner Dense PyC, Density	35 µm, 1.87 g/cc
SiC, Density	35 µm, 3.20 g/cc
Outer Dense PyC, Density	40 µm, 1.83 g/cc
Particle Diameter	820 µm
TRU Heavy Metal Composition	
Np-237	4.10%
Pu-238	1.20%
Pu-239	51.55%
Pu-240	23.88%
Pu-241	7.99%
Pu-242	5.00%
Am-241	5.00%
Am-242M	0.10%
Am-243	1.00%
Cm-242	0.00%
Cm-243	0.00%
Cm-244	0.20%
Cm-245	0.00%
Configuration Temperatures	
Average Thermal Assembly TRU Temperature	770 ^{o}C
Temperature Operating Range	580 ^{o}C to1250^{o}C
Average Graphite Temperature	700 ^{o}C
Average Fast Assembly Fuel Temperature	770 ^{o}C
Thermal Assembly TRU Element Data	
TRU Element Pitch (includes gaps)	36.1 cm
TRU Element Height	79.3 cm
Graphite Block Density	1.74 g/cc
Number of TRU and BP Holes	216
Hole Diameter	1.27 cm
Compact Diameter	1.2446 cm
Coolant Holes	
Number of Inner/Outer Holes	6/102
Diameter Inner/Outer Holes	1.27/1.5875 cm

4.2.4 Physics Analysis

Several block and full transmuter analyses were performed [4.28 and 4.29] to study and to define the impact of the different design parameters on the transmuter performance. The analyses include the TRU material heterogeneity, the burnable poison heterogeneity, the temperature effects, results from different cross-section libraries, different packing factors for the fuel and the burnable poison, burnup analyses, block power distribution, and core power distribution. Some results are presented and discussed in this section.

4.2.4.1 TRU material heterogeneity effect

The block k_∞ values obtained from the second and the third geometrical models relative to the first geometrical models, defined in section 4.2.3, are indicative of the heterogeneity effect. The particle parameters for these analyses are given in Table 4.5. Table 4.6 summarizes the MONK results, which were obtained with the use of the 13193-groups quasi-continuous energy and the 172-groups nuclear data libraries based on JEF2.2. The DRAGON results are also given in Table 4.6.

In these analyses, the block is only loaded with fuel compacts and helium coolant channels. The volume packing fraction for the TRU particles is 0.1238. The heavy-metal loading is 771 grams per block at room temperature. The composition and density of each material in the block are given in Table 4.5. The results show a strong heterogeneity effect of ~14 %, see Table 4.6. The difference in the results between quasi-continuous energy libraries and the multigroup libraries is due to the difference in thermal treatment of the carbon nuclear data. The quasi-continuous energy libraries are lacking the $S(\alpha,\beta)$ treatment for carbon.

Table 4.6. Compact heterogeneity effect

Computer Code	MONK[1]		MONK[2]		DRAGON	
Model	k_∞	$\Delta k_\infty/k_\infty$, %	k_∞	$\Delta k_\infty/k_\infty$, %	k_∞	$\Delta k_\infty/k_\infty$, %
Explicit Modeling	1.2764	--	1.2534	--	1.2539	--
Homogenized Particles	1.1101	-13.02	1.1004	-12.21	--	--
Homogenized compact	1.0928	-14.38	1.0847	-13.46	1.0778	-14.05

1 Quasi-continuous energy library
2 Multigroup library (172 group)

The homogeneous models give inaccurate k_∞ values because these models significantly underestimate the self-shielding of the strong absorption resonances in the plutonium isotopes, particularly Pu-240. This is caused by the fact that the fuel particle dimensions are relatively large compared to the mean free path of neutrons in the low-energy-lying resonances of these isotopes. Because of this effect, the inner zone of the particle is shielded from neutrons by the outer zone and simple homogenization does not account correctly for the self-shielding effect.

The DRAGON results show the same heterogeneity effect similar to MONK. In addition, the results obtained from both codes show an excellent agreement for the explicit and the homogenized models as shown in Table 4.6.

The heterogeneity effect was found to be dependent on the particle composition and the compact packing fraction because of the changes in the neutron spectrum. The difference in k_∞ between the homogeneous and explicit models decreases as the packing fraction increases for fixed particle size or as the fuel radii decreases for fixed packing fraction.

4.2.4.2 Temperature effect

One important feature of this transmuter is the negative temperature coefficient. As the transmuter temperature increases, the neutron spectrum peak shifts toward the absorption resonance of erbium-167 (or other poison material). This results in more neutron absorption in erbium-167. The analysis was performed in steps to define the contribution of each material to this effect using the explicit MONK model with the 172-groups nuclear data library. In this case, the packing fractions are 0.15 and 0.1 for the TRU and the BP, respectively. The first case has

all the materials at 293.16 K. In the second case, the TRU particle temperature was changed to the average operating temperature without changing the temperature of the other materials. The third case is similar to the second case with the graphite temperature of the compact changed to the average operating temperature. The last case changed the graphite block temperature to the operating temperature. Table 4.7 shows the results for these cases. Heating the block materials increases the neutron absorption in erbium-167, which results in a negative temperature coefficient. This enhances the safety performance of the system.

Table 4.7. Temperature effect on the block performance

Burnable Poison Model	K_∞	Relative Difference, %
Cold Conditions	1.1327	
Hot Fuel Particles	1.1112	-1.90
Hot Compact	1.0954	-3.29
Hot Block	1.0562	-6.75

4.2.4.3 Block Sensitivity to TRU and BP Material Loading Parameters

Several parametric studies were performed to characterize the block reactivity. For example, Figure 4.10 shows the block k_∞ as a function of the TRU-particle packing fraction for three different BP packing fractions, at the cold condition. The variation of k_∞ versus the TRU packing fraction shows a peak at low TRU packing fraction. The shift in the neutron spectrum with the packing fraction is responsible for this trend. As the packing fraction decreases, the carbon-to-heavy-metal ratio increases and leads to an increase in the neutron thermalization causing the neutron spectrum to become softer. This result in fewer neutron captures, which enhances the neutron utilization. The improved utilization of neutrons increases Pu-239 fission rate, which enhances the k_∞ as the TRU packing fraction decreases. The packing fraction corresponding to the highest k_∞ differs for the three curves. As the BP loading increases, the spectrum hardens due to the relative neutron absorption increase in Er-167 and Pu-240. Also, less fuel material is required to achieve the same k_∞. The k_∞ increase with the TRU packing fraction, below the peak value of k_∞, is due to the concentration increase of the fissile elements causing more fission reactions.

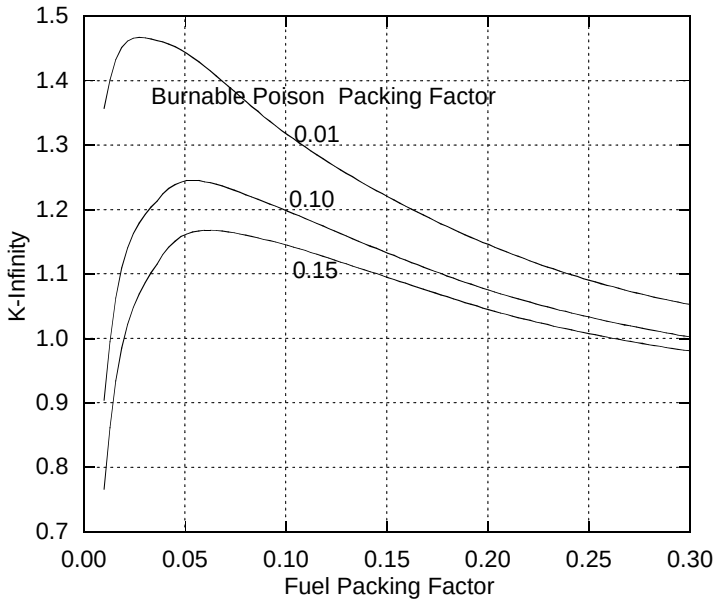

Figure 4.10. Block $K_¥$ as a function of the fuel-packing factor for different burnable poison packing factors

4.2.4.4 Core burnup analysis

MONK burnup calculations were performed for the transmuter using the explicit geometrical model shown in Figure 4.9. The calculations were performed at the average operating temperatures of each material with the 172-groups nuclear cross section library based on JEF2.2. The fuel compact, the graphite block, and the reflector block temperatures are 1043.16, 993.16, and 993.16 K, respectively. The explicit representation of the geometry was maintained in the burnup calculations. A constant power of 600 MW was used in the calculations for 900 days. The packing factors for this configuration are 12.87% and 10% for the TRU and the BP, respectively.

The first step in this analysis was to define the appropriate time step (burnup interval) between subsequent flux calculations because of the large computer time required for each flux calculation. However, the use of a large time step reduces the accuracy of the results. A parametric study was performed to determine the effect of the time steps on the results. Several time steps were used as shown in Figure 4.11. The results show that the fresh transmuter has a K-effective of 1.1005. The K-effective drops to 1.0 after about 500 days. At 900 days, K-effective is very low for this configuration. The burnup parameters converge as the time step is reduced. The results from 12.5 and 25 days time steps are very close. About 44% of the TRU are burned in the first 500 days, and 80% are burned at 900 days. At a K-effective of 0.9, the TRU burnup is 61%. Er-167 is consumed at much faster rate as shown in Figure 4.12. Only the Er-167 isotope is acting as a burnable poison and it is converted to Er-168.

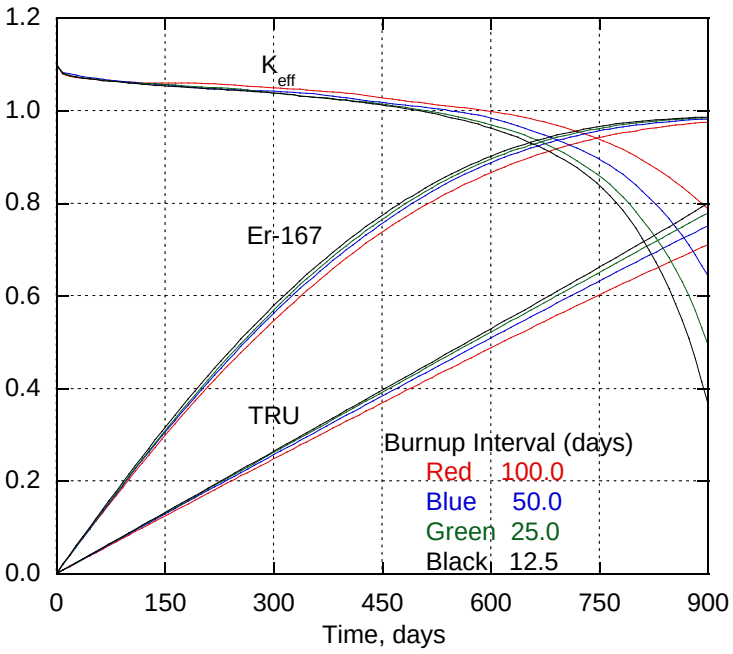

Figure 4.11. Main burnup parameters as a function of the operating time

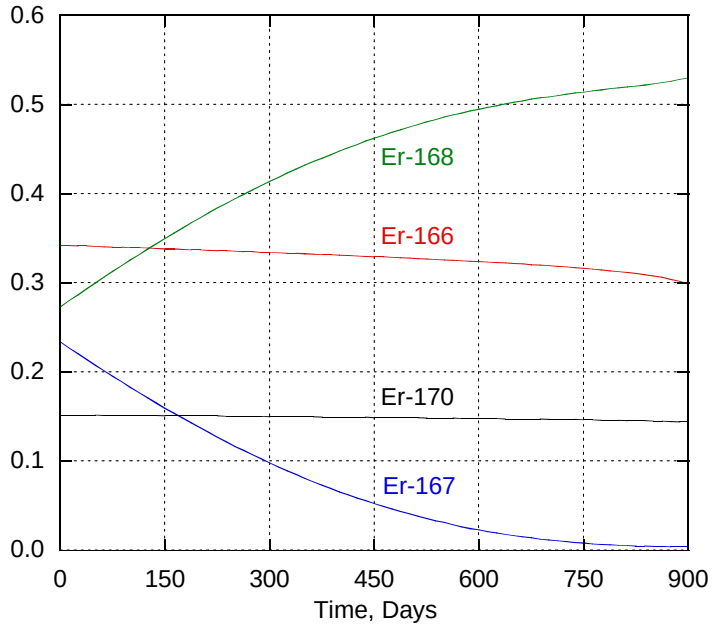

Figure 4.12. Relative atomic concentrations of the erbium isotopes as a function of the operating time

The changes in the atomic concentrations of the different TRU isotopes as a function of the operating time are shown in Figures 4.13 and 4.14. During the critical operation, Pu-239 decreases linearly with the operating time while Pu-240 decreases slowly. In the subcritical operation, the remaining Pu-239 decreases slowly while Pu-240 decreases linearly with the operating time. Pu-241 increases to reach a peak value at about 380 days then it decreases linearly with the operating time. Pu-242 increases slowly during the operation. Pu-237 and Pu-238 decreases slowly during the operation. Am-241 decreases linearly during the operation while Am-243 increases linearly. Am-242m and Cm-244 decrease slowly during the operation.

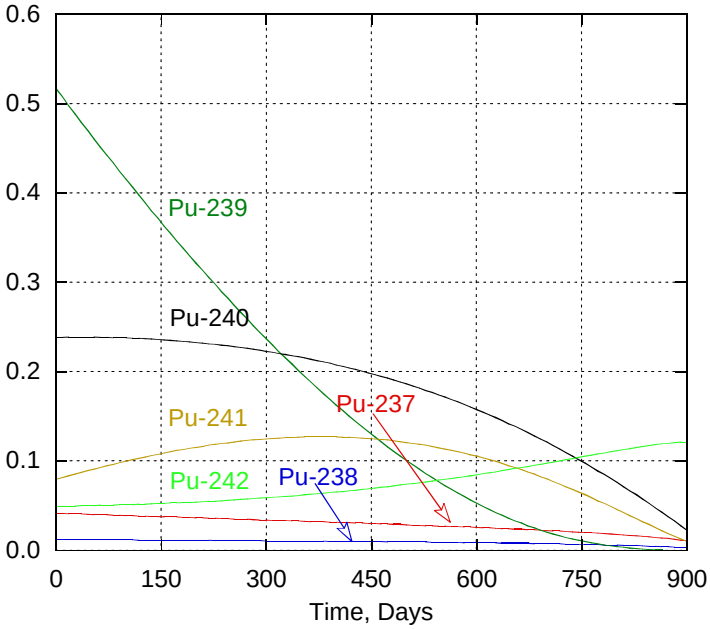

Figure 4.13. Relative atomic concentrations of the plutonium isotopes as a function of the operating time

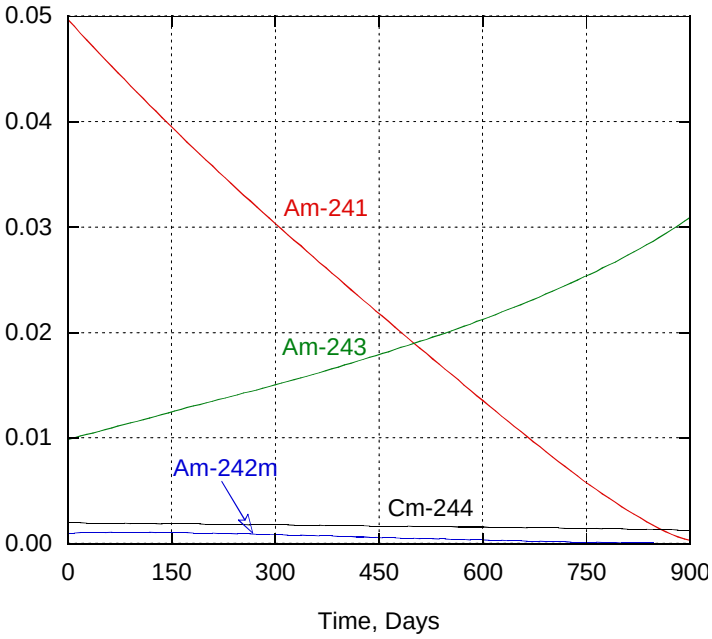

Figure 4.14. Relative atomic concentrations of the americium and curium-244 isotopes as a function of the operating time

4.2.4.5 Transmuter Analysis

As an example, the 3-ring core shown in Figure 4.7 with 36 fuel columns in each of the three core rings is used. The fresh fuel is placed in the middle core ring, away from the reflectors, as shown in Figure 4.7. The block loading with the DF and the TF fuels is shown in Figure 4.15. After one irradiation period that segment was moved to the outer core ring, then, after another irradiation cycle, moved to the inner core ring, then discharged after a third cycle. Thus at any time there are three fuel types in the core with their neutron exposures differing by one cycle. This refueling scheme is referred to as the "3-ring" design. This layout was selected under the assumption that it would maximize the destruction of the TRU waste in the core; however, it was realized that ring-to-ring power peaking would probably require column orificing of the coolant as in the Fort St. Vrain reactor. The LWR TRU waste composition shown in Table 4.8 was used as the "fresh" fuel, and the neptunium and plutonium were put into the DF particles. The americium and curium were loaded into the TF particles, along with an estimate of the discharge DF TRU. The total fuel loading was adjusted to provide a reasonable initial K_{eff}, and the case was run through 5 cycles to obtain an estimate of the actual DF discharge composition. The 5^{th} cycle is subdivided into 24 time steps to increase the accuracy. The TF loading was then adjusted to include the DF TRU discharge, and a true equilibrium cycle was calculated. As part of a sensitivity study, fuel burnups were followed though up to 10 cycles were calculated to reach equilibrium, and the end-of-cycle (EOC) core K_{eff} differed by only 0.7% after about 7 cycles. The average EOC fuel masses differed by only 1 or 2%. A 30 day time step was used in the equilibrium cycle calculations, but going to a 60 day time step did not change the results. 12 month, 18 month, and 24 month refueling intervals were evaluated. The results showed that the 24 month refueling interval (580 day critical cycle at 80% capacity factor) would be difficult to achieve with a "consistent" DF/TF fuel loading in an equilibrium cycle, and the fuel particle fluence limits of $8x10^{25}n/m^2$ would probably be exceeded in this cycle when power peaking effects were included.

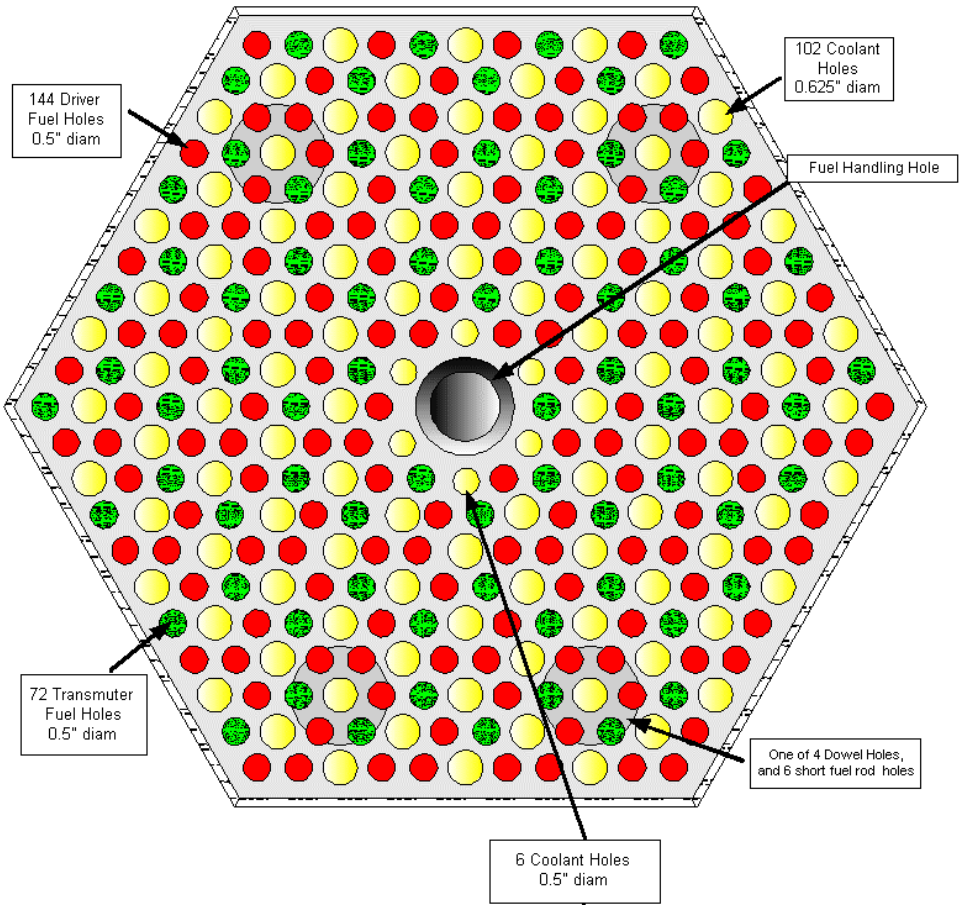

Figure 4.15. Fuel block loading

Table 4.8. Composition (Weight %) of TRU Waste, DF, and TF, for the 18 Month Cycle

Nuclide	LWR TRU Waste	Driver Fuel Particle	Transmuter Fuel Particle
Np-237	4.68%	5.15%	2.46%
Pu-238	1.35%	1.49%	6.58%
Pu-239	51.30%	56.91%	1.18%
Pu-240	20.70%	23.07%	2.66%
Pu-241	7.47%	8.34%	2.27%
Pu-242	4.50%	5.05%	29.5%
Am-241	8.18%	0%	27.5%
Am-242m	0.03%	0%	0.1%
Am-243	1.48%	0%	11.7%
Cm-242	0.0%	0%	0.39%
Cm-243	0.0%	0%	0.02%
Cm-244	0.29%	0%	14.99%
Cm-245	0.02%	0%	0.58%
Total	100.0%	100.0%	100.0%

The fuel compositions are given in Table 4.8 and the particle parameters for both the DF and the TF are given in Table 4.9. The results from the analysis with an 18 month fuel cycle are

summarized in Table 4.10. In this case 75.2% total transmutation of the input TRU waste was achieved, along with 98.6% destruction of the Pu-239 and 94.5% destruction of the Pu-241. The DF average fluence was 7.7×10^{25} n/m^2 ($E \geq 0.18$MeV), which is below the 8×10^{25} n/m^2 limit. The TF particles reached 5.3×10^{25} n/m^2 average fluence. These results show very high burnup for this fuel cycle; however, the ring power variations were unacceptable, with a factor of 5 difference between the fresh and oldest fuel rings.

Table 4.9. Parameters of the fuel particles

Particle Parameter	Dimension in μm (microns)		Density in g/cm^3		Material
	Driver Fuel Particle	Transmuter Fuel Particle	Driver Fuel Particle	Transmuter Fuel Particle	
Kernel diameter	300	250	10.36	5.00	$MO_{1.7}$
Coating thickness:					
Buffer	150	100	1.00	1.00	C
Inner pyrolitic	35	35	1.85	1.85	C
Silicon carbide	35	35	3.20	3.20	SiC
Outer pyrolitic	40	40	1.85	1.85	C

A different distribution of the three fuel ages in the core was considered, termed the "Distributed Core" design, in which the columns of each segment are more evenly distributed throughout the core. The idea was to minimize ring power peaking and avoid the need to move all the fuel in the core at each reload, since fuel of a given age would remain in a fixed position in the core throughout its life. The results show that the total TRU destruction was not significantly reduced from the 3-ring case, however, the maximum burnup and fluence is slightly lower.

The fuel isotopic changes for the three-ring core with 24 month fuel cycle are shown in Figure 4.16 for both fuel types. It is clear that this transmuter build Cm isotopes that require fast spectrum for destruction.

Table 4.10. Summary of results for 3-ring, 18 month deep-burn cycle, physics study

Parameter	Value
▪ Fuel Cycle Length (months/days)	18/540
▪ Segment TRU input (kgs)	558
▪ Segment Irradiation time (years/cycles)	4.5/3
▪ Total TRU destruction (%)	80%
▪ Pu-239 destruction (%)	99%
▪ Pu-241 destruction (%)	94%
▪ Driver Fuel (DF):	
▪ DF load (kgs)	502.5
▪ Burnup (GWD/MT)	500
▪ Fluence (n/cm^2 for E≥ 0.18MeV)	5.5×10^{21}
▪ Transmuter Fuel (TF):	
▪ TF Load (kgs)	110.9
▪ Burnup (GWD/MT)	112
▪ Fluence (n/cm^2 for E≥ 0.18MeV)	6.2×10^{21}

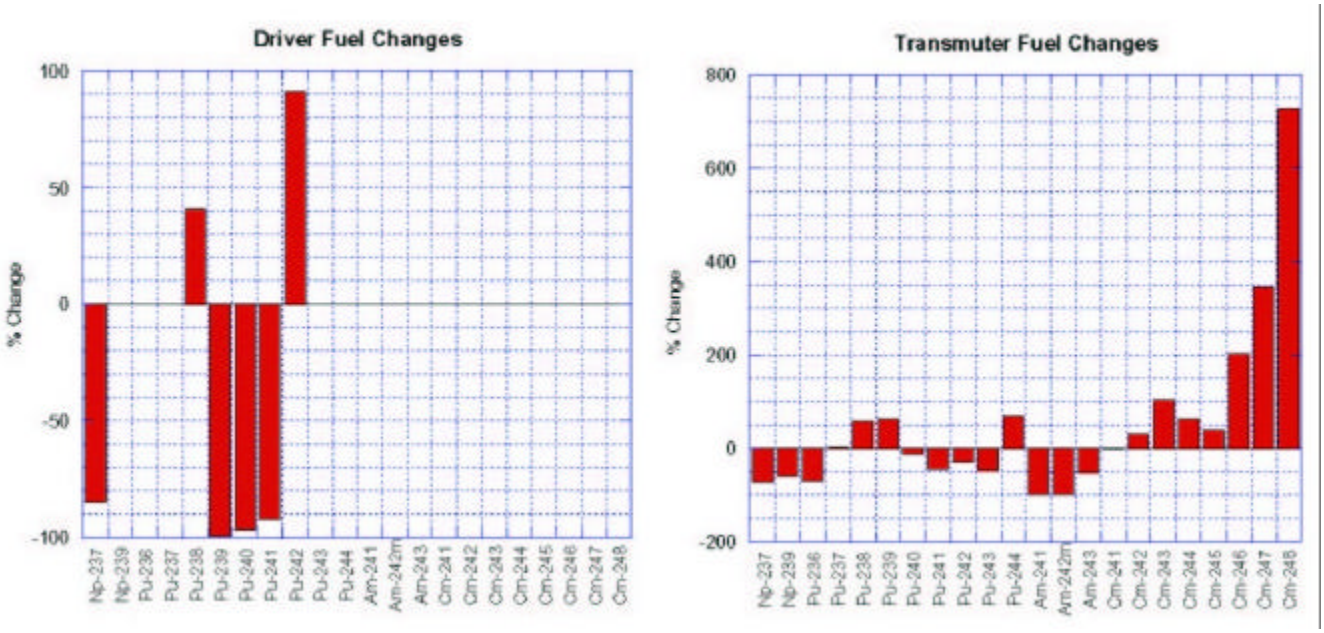

Figure 4.16. Fuel isotopic changes for 3-ring, 24 month fuel cycle

4.2.5. Conclusions

Several conclusions were obtained from the presented results. First, accurate geometrical models for Monte Carlo analyses were developed for the block and the whole transmuter based on the high temperature gas reactor technology with TRU in the form of coated particles. Second, these models were used successfully to perform detailed physics analyses including burnup. Third, the results show the need for using detailed geometrical models including explicit presentation for the multi-layers fuel particles because material homogenization introduces significant errors in the performance parameters. Fourth, the parametric studies show the potential for adjusting the block design to optimize the transmuter performance and the fuel cycle for achieving high transmutation rate.

The presented transmuter analyses show that the high temperature gas cooled reactor technology can be used to destroy TRU waste using ceramic coated, TRISO, fuel particles. The burnup and fluence limits of these particles containing TRU needs to be demonstrated by test, but with the current limits, over 70% of the TRU waste could be destroyed in a single irradiation cycle. This cycle also eliminates all the fissile plutonium in the waste, and effectively extracts more than 20% extra energy from the original LWR fuel. Since the gas-cooled reactor operates safely at high coolant temperatures, this energy can be converted to electricity at ~50% efficiency, or used to generate hydrogen or process heat, clearly improving the economics of any transmuter system.

The analysis also indicated that the transmuter parameters can be optimized to avoid power peaking. The radial zoning can be used to limit power peaking to acceptable levels. The system should have a negative temperature coefficient for safe operation, though further analysis is required to confirm this. The use of two fuel types, a driver and a transmuter particle, with a single extra reprocessing step to use the driver fuel discharge transuranics, maximizes the TRU waste destruction. The results show that 75% to 82% of the TRU waste can be transmuted, and 94% to 98% of the fissile plutonium can be destroyed. If the development and qualification of the TF (containing americium and curium) is delayed, a transmutation cycle could be operated using only the DF, plus a burnable poison such as erbium for reactivity control.

Further studies to analyze the proliferation issues of the Pu + Np production line, to confirm the driver fuel form with neptunium, to study the transmuter fuel form and fabrication issues, to define the driver fuel reprocessing method for the TRISO fuel form, to study the adaptation of the discharged transmuter fuel for the repository or the fast transmuter, and to characterize the fuel particle performance for high burnup are warranted.

4.3 Transuranic Management in a Fast Reactor Closed Fuel Cycle

An intense, sustained neutron flux is required to fission significant amounts of transuranic (TRU) material, and a variety of possible neutron sources (e.g., a nuclear reactor, accelerators with spallation targets, etc.) have been proposed. The primary advantage of nuclear reactor systems is the demonstrated technology of fission reactors coupled to an efficient secondary system for converting fission energy into reactor power. *Any fission-based destruction system "burns" TRU at the same rate, ~1 MWt-day per gram fissioned.* However, proposed reactor systems have a wide variety of transmutation characteristics and fuel cycle strategies.

Fast Spectrum Effects and Proliferation Resistant Features

The hard neutron energy spectrum of fast reactor systems leads to several favorable effects for TRU management. First, actinides are preferentially fission, not converted to still higher actinides, because the fission-to-capture ratio is much higher as shown in Fig. 4.17. This implies that fast systems are more "efficient" in destroying actinides because fewer neutrons are lost to capture reactions before eventual fission. Furthermore, the generation of higher actinides (which can be problematic for fuel handling) is suppressed.

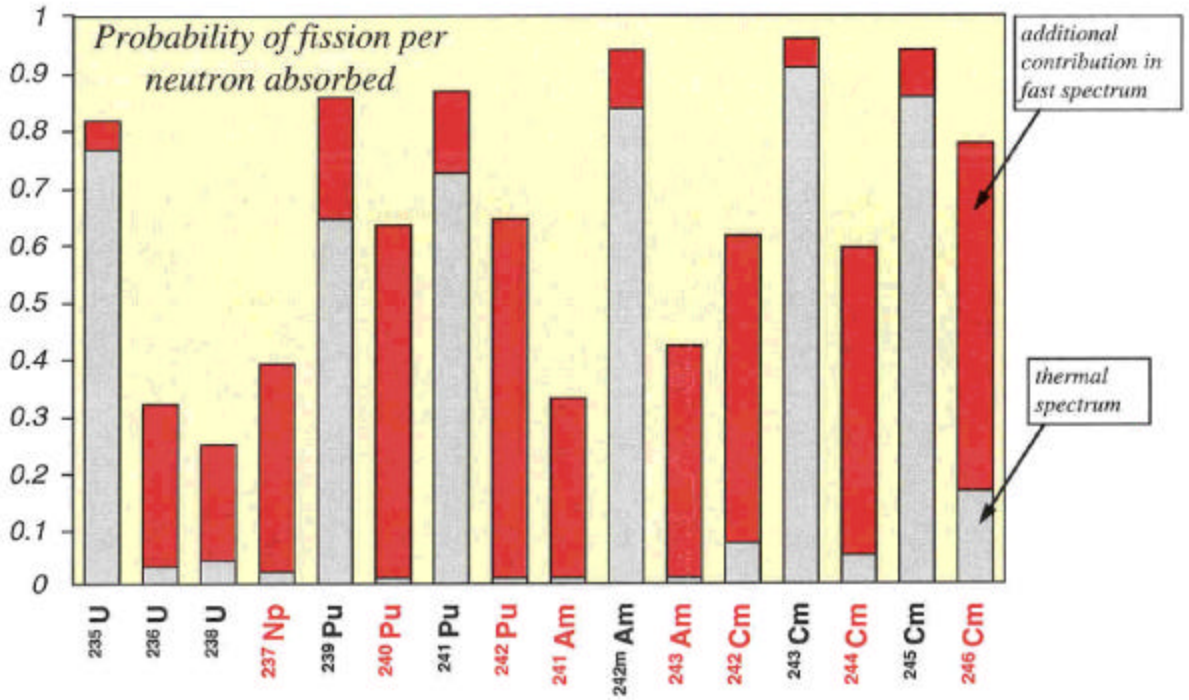

Figure 4.17. Comparison of Fast and Thermal Spectrum Fission/Absorption Ratio

Because the capture cross section of the fission products also decreases sharply at high energies, the performance penalties for the unavoidable carryover of recycle fission products are minor. For example, the core performance impact for incomplete separation of the rare-earth fission products (up to 60% recycled with TRU) are shown in Table 4.11. As the impurity concentration increases, the burnup reactivity loss increases slightly indicating a lower internal breeding ratio (neutrons lost to parasitic capture). However, the overall effect is quite small with

only a 0.08% ?k reactivity swing increase at 25% rare earth recycle. The potential impact of significant rare-earth concentrations on the fuel properties and irradiation performance will more likely limit the desired fission product separations purity.

Table 4.11. Core Performance Impact of Rare-Earth Recycle Carry-Over

Rare-Earth Carry-over Fraction, %	0	9.1	11.1	25	60
Burnup Reactivity Loss, %?k	-0.30	-0.29	-0.27	-0.22	+0.11
Mass Flows, kg/y					
Fission Products	0	11	14	38	167
Fissile TRU	483	483	484	484	492
Total Heavy Metal	6700	6690	6680	6660	6530
Total Breeding Ratio	1.174	1.173	1.171	1.166	1.136

Thus, a fast spectrum system can be utilized for repeated recycle without concern for the neutronics effect of fission product carry-over nor the buildup of higher actinides (as observed for thermal spectrum recycle). This approach has been pursued in a variety of international programs including the French CAPRA project and the U.S. Integral Fast Reactor (IFR) fuel cycle. The closed fuel cycle based on pyrometallurgical processing and injection casting re-fabrication offers some proliferation advantages. This technology can be deployed on a small-scale with a compact fuel cycle facility collocated with the reactor plant; this eliminates off-site transport of the fuel and recycle materials.

In addition, the separation process is inherently incomplete with no separation of the TRU elements (e.g., no pure plutonium stream), partial separation from the bulk uranium, and partial carry-over of rare-earth fission products. This requires remote fuel processing and refabrication. To this end, an injection casting process for metal fuel with loose dimensional tolerances allowed by sodium bonding has been developed.

Fuel Management Strategies – Burner Reactors

All conventional reactor systems utilize fuel that is primarily uranium; thus, the destruction of plutonium is at least partially compensated by in-situ production of Pu-239. In addition, “once-through” fuel management strategies are commonly employed, and the majority of the plutonium never gets destroyed – it simply ends up in the spent fuel. However, in a closed fuel cycle, repeated recycles are employed to achieve extensive fission of the transuranics. As illustrated in recent fuel cycle studies, it is vital to maintain low processing fractions to ensure containment of the TRU during this destruction campaign [4-30].

The wide range of conversion characteristics achievable in a fast spectrum system provides for flexibility in TRU management strategy. The neutron balance in a fast spectrum allows a net production of fissile material; and the traditional approach employed excess fertile material to breed additional TRUs. These fast breeder reactor configurations were envisioned for a rapidly expanding nuclear power economy where fissile material was scarce and expensive. Conversely, the fast reactor can readily be configured to achieve a net destruction (burning) of TRUs. Given the current status of the nuclear fuel cycle with stockpiles of excess weapons material, separated civil plutonium, and TRU-containing spent fuel, fast burner reactor configurations have been targeted in recent studies to reduce the global TRU inventory.

In a standalone mode, a fast burner reactor can reduce existing stockpiles of TRU material. Alternately, fast burner systems can be utilized to complement a sustained nuclear power capability using once-through (enriched uranium) systems that inherently produce TRUs. As shown in the recent AAA multiple strata study [4.30], a limited capacity of fast burner systems can be utilized to stabilize the total TRU inventory. This approach may be advantageous as it allows

exploitation of current enriched uranium resources without continued buildup of once-through fuel cycle wastes. For either of these missions (stockpile reduction or waste transmutation), high net TRU consumption rates are advantageous; the number of systems required to complete the mission will be directly dictated by the consumption rate.

To achieve net consumption of TRU, their fission rate must exceed the production rate of new TRU. The conversion ratio is a useful measure of the relative rates, defined as

$$TRU\ conversion\ ratio\ (CR) = \frac{TRU\ \ production\ rate}{TRU\ destruction\ (\ fission)\ rate}$$

Thus, CR=1 denotes a TRU sustaining system, and low conversion rates are targeted for fast burner systems. It is important to reiterate that the fission of actinides (regardless the reactor type) results in ~1 MWt-day of energy per gram destroyed. Assuming that TRUs are the dominant fission isotope, this implies that the net TRU destruction is (1-CR) grams per MWt-day of energy produced by the reactor system.

Fast burner reactor designs have been developed for a variety of missions. In Ref. 2 [4.31], burner configurations were developed for weapons plutonium disposition. In particular, a moderate burner design (CR~1/2 using conventional fuel forms) and a pure burner design (CR=0, using nonuranium fuel form) were developed. In that particular study, a fixed poison (hafnium) was added to the pure burner fuel to improve some performance aspects; and the TRU consumption performance at startup and with recycle was investigated in detail as summarized in Table 4.12. In previous work (Ref. 4.32), pure burner designs without such additives were also considered; relevant results are also shown in Table 4.12. As expected the pure burner designs (no uranium) achieve the maximum destruction rate of 1.0 g/MWtd, while the moderate burner (CR=0.6) achieves a destruction rate of roughly 0.4 g/MWtd.

The reduced fertile material loading results in a significant increase in the reactivity loss rate; for example, the conventional system has a burnup swing less than 1% for a two year cycle compared to a 12% reactivity loss for the short cycle TRU burner. Thus, *burner designs will have a much greater reliance on reactivity compensation systems* than conventional fast reactors; this also implies that beginning of cycle (BOC) excess reactivity requirements will be higher.

Table 4.12. Comparison of Fast Burner Reactor Performance (from Refs. 4.31 and 4.32)

Parameter	Conventional Design[3]	Moderate Burner[3]	Pure Burner[3]	TRU Burner[4]	MA Burner[4]
Conversion Ratio	1.19	0.59	0	0	0
Power, MWt	840	840	840	1200	1200
Cycle Length, months	23	12	12	8.3	15
Capacity Factor, %	85	85	75	80	80
TRU Enrichment, wt%TRU/HM	20	19/23	100	100	100
Net TRU destruction rate, kg/y	-41	95	231	350	350
Normalized TRU destruction, g/MWtd	-0.157	0.365	**1.005**	0.999	0.999
Burnup Swing, %?k	**0.73**	3.45	3.93	**12.1**	4.33
Delayed Neutron Fraction, ß$_{eff}$	3.44E-3	3.16E-3	1.97E-3	2.39E-3	**1.49E-3**
Doppler coefficient, cents/K	-0.14	-0.11	**-0.02**	-	-
Total Sodium Void Worth, $	6.32	1.72	**18.3**	3.33	**22.8**

Several of the key reactivity feedback coefficients for the various burner configurations are also summarized in Table 4.12. The suppression of U-238 fission results in a significant reduction in the delayed neutron fraction; a delayed neutron fraction of 0.002 is characteristic of plutonium fission. This reduction is particularly severe (0.0015) for the minor actinide (MA) pure burner where plutonium fission is also eliminated. In addition, the removal of U-238 eliminates the dominant Doppler reactivity feedback mechanism; and the pure burner designs exhibit a near-zero Doppler coefficient. Finally, large variations in the sodium void worth are observed between the burner configurations, ranging from $1.7 to $23. Thus, one must be careful to design the burner system in a manner that does not exacerbate the void worth.

It was concluded in Ref. 4.31 that moderate burner designs (using conventional fuel) can be utilized without adversely impacting fast reactor performance. However, pure burner designs exhibit several undesirable reactivity feedback features. Thus, they were deemed an unattractive short-term option because of the development program needed to refine the system performance and develop the new fuel form. The work in Ref. 4.31 is typical of fast burner design studies conducted worldwide. In European and Japanese studies, separate burning of minor actinides is also considered as they have been segregated by PUREX processing. The general performance trends noted above are confirmed in diverse international results.

Another important consideration is the impact of alternative TRU feed streams on the system performance. For conventional designs (CR=1), only the startup performance will be impacted because no additional TRU material is required. However, for burner designs external TRU is required for both startup material and makeup feed with recycle. The time dependence of the TRU isotopic inventory was evaluated in Ref. 4.31 in detail. In general, the makeup feed composition becomes more important at low conversion ratios, and a buildup of fertile isotopes (low cross section) with recycle is observed. Performance of a moderate burner core with TRU feeds based on LWR spent fuel, weapons plutonium, and LMR recycle are compared in Table 4.13.

Table 4.13. Comparison of Alternative Feed Burner Reactor Performance (from Ref. 4.31)

Feed Stream	Weapons Plutonium	Recycled LMR	LWR Spent Fuel
Conversion Ratio	0.54	0.51	0.49
TRU Enrichment, wt%TRU/HM	23/29	26/33	29/36
Net TRU destruction rate, kg/y	110	117	124
Normalized TRU destruction, g/MWtd	0.448	0.477	0.505
Burnup Swing, %?k	2.90	2.58	2.37
Delayed Neutron Fraction, ß$_{eff}$	3.12E-2	3.09E-3	3.04E-3
Doppler coefficient, cents/K	-0.09	-0.08	-0.07
Total Sodium Void Worth, $	2.94	3.79	4.61

The alternative feed comparisons indicate that the overall performance of the conventional burner design with recycle is similar for a wide variety of potential feeds. The primary performance changes are caused by differences in the enrichment requirement dictated by the fissile content of the feed material. Feeds with a lower fissile content (e.g., LWR TRU) increase the driver enrichment and TRU loading requirements, but the higher Pu-240 content reduces the burnup reactivity loss rate. In addition, the higher Pu-240 content will increase the sodium void worth. More severe effects have been observed for fuel cycles with a radical change in the fissile

content (e.g., pure minor actinide burner). Although fissile material can be readily generated with recycle; criticality can be quite difficult to achieve for the startup core.

4.4 Differences between Fast Reactor and ADS Safety Behavior

Traditional safety performance requirements for nuclear reactor have been developed for critical systems, whose kinetics characteristics differ significantly from sub-critical, accelerator-driven systems. In a critical reactor, relatively small amounts of reactivity (negative or positive) can produce large changes in the fission rate. This implies that reactor systems are sensitive to feedback effects (e.g., temperature-induced reactivity changes), which have been exploited to enhance the passive safety of advanced reactor concepts. In contrast, highly subcritical systems are completely source-dependent, with little response to small reactivity changes. As the multiplication factor increases, the system becomes more sensitive to reactivity changes particularly near the critical state.

The overall safety performance of subcritical systems should not be characterized as obviously worse or better than advanced reactor systems. The transient behavior of subcritical systems indicates a different set of inherent strengths and weaknesses. Reactivity-induced transients are particularly benign in subcritical systems, because the system power is insensitive to the reactivity insertion. However, response to loss-of-cooling events is more favorable in critical systems which passively shutdown based on temperature feedbacks; whereas, an active shutdown device may be required for the subcritical system.

In general, subcritical systems are less responsive to positive reactivity insertions than critical reactors. However, this implies that larger negative reactivity insertions are needed to shut down the systems if the source remains. Thus, conventional (reactor) control systems which rely on small reactivity changes are not readily adapted to ADS, and alternative control strategies are being investigated.

References

4.1 Energy Information Administration, http://www.eia.doe.gov.

4.2 Nuclear Energy Agency Working Group, *Plutonium Fuel: An Assessment*, NEA-OECD Report (1989).

4.3 M. Salvatores, Private Communication, 2001.

4.4 *Physics of Plutonium Recycling: Volume 1 – Issues and Perspectives*, NEA-OECD Report (1995).

4.5 G. Youinou, *et al, Heterogeneous Assembly for Plutonium Multi-recycling in PWRs: The CORAIL Concept*, GLOBAL 2001.

4.6 T. K. Kim, *Assessment of CORAIL-Pu Mutli-Recycling in PWRs*, Argonne National Laboratory, ANL-AAA-018 (2002).

4.7 T. K. Kim, *et al*, *Assessment of TRU Stabilization in PWRs*, Argonne National Laboratory, ANL-AAA-020 (2002).

4.8 S. Aniel*, et al, Plutonium Recycling in PWR: The CORAIL Concept,* ICONE-8: Eight International Conference on Nuclear Engineering (2000).

4.9 Fuel Review: Design Data, *Nuclear Engineering International.*

4.10 *WIMS – A Modular Scheme for Neutronics Calculations*, AEA Technology.

4.11 J. A. Stillman and R. N. Hill, *ADS Transmuter Performance with Minor Actinide External Feed,* Argonne National Laboratory, ANL-AAA-024 (2002).

4.12 1990 Recommendations of the International Commission on Radiological Protection, ICRP Publication 60, Annals of the ICRP, Vol. 21, Nos. 1-3, Pergamon Press, Oxford (1991).

4.13 R. N. Hill, et al, Multiple Tier Fuel Cycle Studies for Waste Transmutation, ICONE-10: Tenth International Conference on Nuclear Engineering (2002).

4.14 H.J. Rütten, compiler, “Potential of Thorium-based Fuel Cycles to Constrain Plutonium and to Reduce the Long-Lived Waste Toxicity,” Draft Final Report on the Co-ordinated Research Programme, Reg. No. 13.30.08, International Atomic Energy Agency, July 2000.

4.15 H. Gruppelaar and J. P. Schapira, editors,"Thorium as a Waste Management Option", Project report: Nuclear Science and Technology, EUR 19142EN, 2000.

4.16 A. Galperin, M. Segev, and M. Todosow, "A Pressurized Water Reactor Plutonium Incinerator Based on Thorium Fuel and Seed-Blanket Geometry*", Nucl Tech, Vol. 132*, Nov. 2000.

4.17 H-K. Joo and Y-J. Kim, “Potential of Thorium-based Fuel Cycle for 900 Mwe PWR Core to Constrain Pu and to Reduce Long-term Toxicity,” in Reference 1.

4.18 D. Biswas et al., “Weapons-Grade Plutonium Disposition in Pressurized Water Reactors,” *Nucl. Sci. & Engr., 121*, 1, 1995.

4.19 A. Galperin et al., Washington ANS, Nov. 2000.

4.20 A. Baxter and M. Fikani, “Reactor-Based Transmutation - Physics Studies of the Gas-Cooled, Graphite-Moderated, Deep-Burn Reactor,” General Atomic Report GA-501-0-TRT-000140, June 30, 2002.

4.21 The ANSWERS Software Package, MONK - A Monte Carlo Program for Nuclear Criticality Safety and Reactor Physics Analyses, User Guide for Version 8, ANSWERS/MONK(98)6, AEA Technology, UK.

4.22 G. Marleau et al., “A User’s Guide for DRAGON,” IGE-174, Rev. 3, Ecole Polytechnique de Montreal (Dec. 1997).

4.23 K. L. Derstine, “DIF3D: A Code to Solve One-, Two-, and Three-Dimensional Diffusion Theory Problems,” ANL-82-64, Argonne National Laboratory, (1984).

4.24 W. S. Yang and H. Khalil, “Analysis of the ATW Fuel Cycle Using the REBUS-3 Code System,” *Trans. Am. Nucl. Soc.,* 81, 277 (1999).

4.25 H. R. Trellue and D.I. Poston “User’s Manual, Version 2.0 for Monteburns, Version 5B”, LANL LA-UR-99-4999, 1999.

4.26 J. F. Briesmeister, ed., "MCNP A General Monte Carlo N-particle Transport Code Version 4B," Los Alamos National Laboratory, LA-12625-M Version 4B (1997).

4.27 A. G. Croff, "A User's Manual for ORIGEN2 Computer Code," ORNL/TM-7175, Oak Ridge National Laboratory (July 1980).

4.28 Yousry Gohar, Temitope A. Taiwo*, and Phillip J. Finck, “Neutronics Verification of GA Accelerator Transmutation of Waste Concept,” Argonne National Laboratory Report, ANL/TD/TM 01-17, 2001.

4.29 Y. Gohar, T. A. Taiwo, and P. J. Finck, “Transmutation of Light Water Reactor-Discharge-Transuranics in a High Temperature Gas Cooled Reactor,” Accelerator Applications/Accelerator Driven Transmutation Technology and Applications 01 (AccApp-ADTTA’01), Reno, Nevada, November 2001.

4.30 R. N. Hill, T. A. Taiwo, J. A. Stillman, D. J. Graziano, D. R. Bennett, H. Trellue, M. Todosow, W. G. Halsey, and A. Baxter, “Multiple Tier Fuel Cycle Studies for Waste Transmutation,” *Proc. of ICONE-10*, April 14-18, 2002.

4.31 R.N. Hill, D. C. Wade, J. R. Liaw, and E. K. Fujita, “Physics Studies of Weapons Plutonium Disposition in the Integral Fast Reactor Closed Fuel Cycle,” *Nucl. Sci. Eng.*, 121, 17 (1995).

4.32 R. N. Hill, “Evaluation of Reactivity Coefficients for Transuranic Burning Fast-Reactor Designs,” invited, *Trans. of the ANS*, 65, 450 (1992).

5. Integrated system performance studies

As has been mentioned earlier, the interaction between the two components of a dual tier system is not yet fully understood; in particular their optimum combination (or, equivalently, the optimum degree of Plutonium burning in the first Tier) needs to be better understood. Section 5.1 summarizes some recent findings in this area.

Several recent studies [5.1, 5.2] have attempted to compare the performances of various systems for transmuting waste. The comparison was generally performed for global transmutation criteria (this can be measured by the time needed for the residual radiotoxicity to decay to uranium ore levels) and other criteria of interest (for example: net cost, volume of high level waste). Global findings of these studies are summarized in Section 5.2.

5.1 Impact of Tier 1 Partial Burnup on Tier 2 Fast System Performance

A variety of multiple tier fuel cycle strategies have been conceived where the TRU are partially consumed in commercial systems, with the remaining material passed on to a dedicated fast spectrum burner. This approach is intended to provide a more cost effective path for transmuting the waste material, particularly early in the process when the fissile content is still high. However if the initial irradiation is conducted in a thermal spectrum system, the fissile materials will be preferentially consumed. Furthermore, significant quantities of higher actinides (americium and curium) may be generated, particularly with deep burnup. Thus, the feed material for the Tier 2 fast spectrum system is less reactive but highly radioactive; and the final destruction of the radiotoxic minor actinides is more difficult.

For example, the makeup feed for a second tier accelerator driven system (ADS) based on the combination of the minor actinides (Np, Am, and Cm) which bypass the first tier and 50% burnup first tier discharge is compared to discharge LWR isotopics in Table 5.1. This mixed feed is quite degraded from the initial commercial ALWR discharge; the fissile plutonium fraction has decreased from 56% to 32% with buildup of higher actinides.

Table 5.1. LMR Isotopics for Case 3m and Second Tier Case 1z

Nuclide	ALWR 10 y cool	Case 3M Eq. Charge	Case 3M Eq. Disch.	Case 1Z ALWR Disc.	Case 1Z Eq. Charge	Case 1Z Eq. Disch.
Np-237	6.6	3.8	2.6	0.0	5.9	4.1
Pu-238	2.7	5.6	6.5	5.0	7.9	8.8
Pu-239	48.7	26.0	16.7	14.6	**6.7**	5.4
Pu-240	23.0	33.2	37.1	35.7	28.7	28.6
Pu-241	6.9	6.6	7.2	**17.7**	7.4	6.3
Pu-242	5.0	10.8	13.2	16.5	18.2	19.8
Am-241	4.7	4.5	3.8	3.2	7.3	5.6
Am-242M	0.0	0.2	0.3	0.0	0.4	0.5
Am-243	1.5	3.7	4.6	4.0	7.6	8.2
Cm-242	0.0	0.0	0.4	0.0	0.0	0.5
Cm-243	0.0	0.0	0.0	0.0	0.0	0.0
Cm-244	0.5	2.7	3.9	**2.6**	5.8	7.2
Cm-245	0.0	0.8	1.1	0.4	1.6	2.0
Cm-246	0.0	0.4	0.6	0.0	0.9	1.1
Fissile Pu	55.6	32.6	23.9	32.3	**14.0**	11.6
Np-Chain	18.3	15.7	14.6	21.3	22.2	18.0

The significant change in feed isotopics between the ALWR discharge and mixed Tier 1 discharge impacts the performance of the second tier system. The reduced fissile content of the heavy metal requires a significant increase in the TRU inventory; the resulting change in core performance parameters is shown in Table 5.2. The TRU inventory increases by 35% compared to the direct fast system case; and this results in a corresponding 25% in the average discharge burnup. Because the systems exhibit similar discharge fluence levels, this *reduced burnup can not be recouped* by increasing the fuel residence time. The decreased fissile content (more fertile material) results in lower burnup reactivity loss rates. A significant reduction in burnup swing from 4.1 to 2.5 %?k is observed.

Table 5.2. Subcritical Fast System Performance

Parameter		ALWR Feed	Tier 1 Feed
BOEC Heavy metal inventory (kg)		2708	**3657**
Fuel enrichment (weight % TRU in matrix)	Inner Zone	59	61
	Outer Zone	70	72
Multiplication Factor	BOEC	0.969	0.970
	EOEC	0.928	0.945
Cycle Length, days		140	140
Burnup reactivity loss (%?k)		4.14	**2.52**
Peak linear power (W/cm)		385	361
Discharge burnup (MWd/kg)		273	**209**
Peak fast fluence (10^{23} n/cm^2)		3.73	3.70

5.2 Summary of recent integrated system performance studies

Previous sections have demonstrated the individual performances of various systems for transmuting nuclear waste. Essentially, thermal reactors can do an excellent job at transmuting plutonium and neptunium, and under certain circumstances can help reduce the inventories of Americium. Various schemes are being proposed which a significant spectrum in R&D requirements. Fast reactors or accelerator driven systems on the other hand can readily destroy all transuranics but still require significant investment for fully closing the fuel cycle. The question we would like to address here is: as far as transmutation performance is compared, are there fundamental differences between various schemes based either on single or multiple tier approaches? (Other questions will of course quickly arise, such as: which of these systems is the cheapest, or fastest, or more robust, to implement? These questions were handled in the studies presented here, and will be briefly discussed).

The results of two major recent studies can be used:

- A comparative study by the OECD on “Accelerator-driven Systems (ADS) and Fast Reactors” [5.1], which compares several conventional transmutation scenarios.
- A comparative study by the US Advanced Accelerator Applications (AAA) program on “Candidate Approaches for an Integrated Nuclear Waste Management Strategy- Scoping Evaluations” [5.2], which compares other slightly less conventional options.

The OECD study compared six representative fuel cycles described in Figure 5.1. Ignoring the Pu Burning cycle which is only a partial transmutation cycle, they are:

- The LWR once through cycle as currently implemented in many countries, where the SNF will eventually be sent to a geological repository;
- TRU burning in an advanced critical fast reactor optimized for a low conversion ratio (CR = .5)

- TRU burning in an ADS (CR = 0)
- The double strata approach with LWR's and fast reactors in the first stratum and ADS's MA burners in the second stratum;
- The pure fast reactor strategy based on the IFR concept where the fuel cycle is closed for all actinides, representing the long term goal for nuclear development.

Note that for all fast reactor and ADS cases, multirecycling of the fuel was assumed.

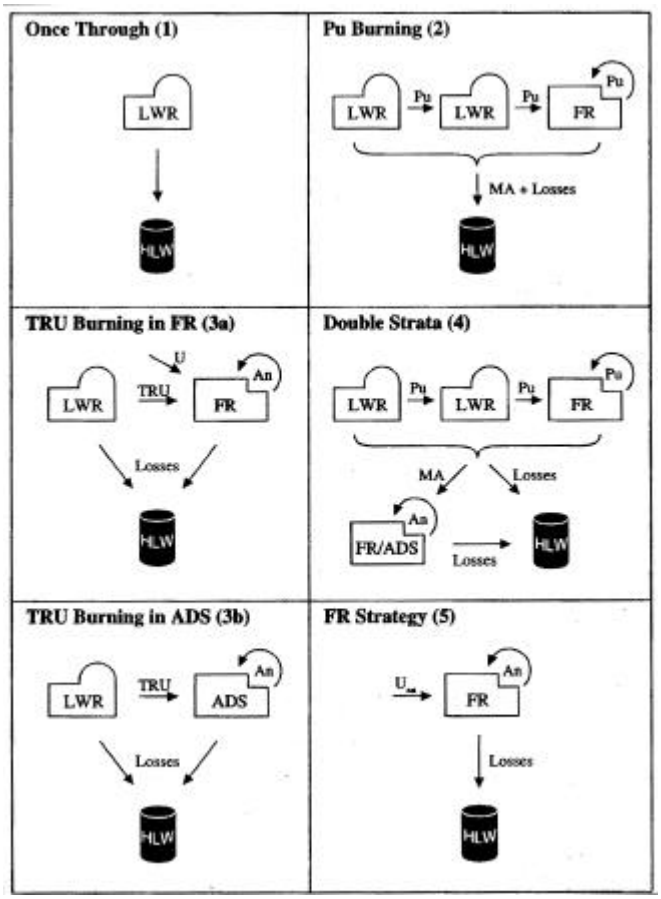

Figure 5.1. Overview of principal fuel cycle schemes

These cases were analyzed with consistent neutronics methodologies and used generally accepted assumptions for the performances of fuels and separation processes. In particular, the loss rate during each fabrication and separation pass was assumed to be .1% of the throughput.

Main results are given below:

- Figure 5.2 provides the electricity contribution of each reactor type to total production and indicates that the "TRU burning in ADS" case requires the smallest additional (Non LWR) infrastructure of all cases.
- Figure 5.3 provides the resource efficiency and High Level Waste (HLW) production relative to the open fuel cycle. It indicates that all cycles generate roughly the same amount of HLW, and use the same amount of natural resources. As expected, the fast reactor case requires a significantly smaller amount of natural uranium.
- Figure 5.4 provides a preliminary estimate of the relative costs of the various scenarios, and shows that within large uncertainties the TRU burning in fast reactors and the double strata approaches appear to be the most attractive.

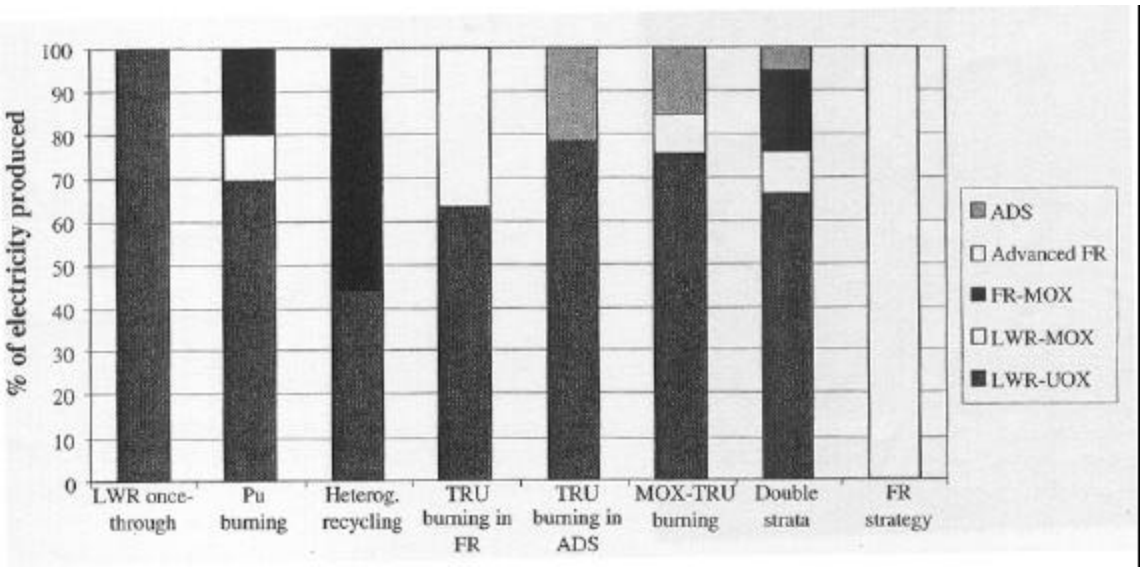

Figure 5.2. Electricity contributions of reactor components for different schemes

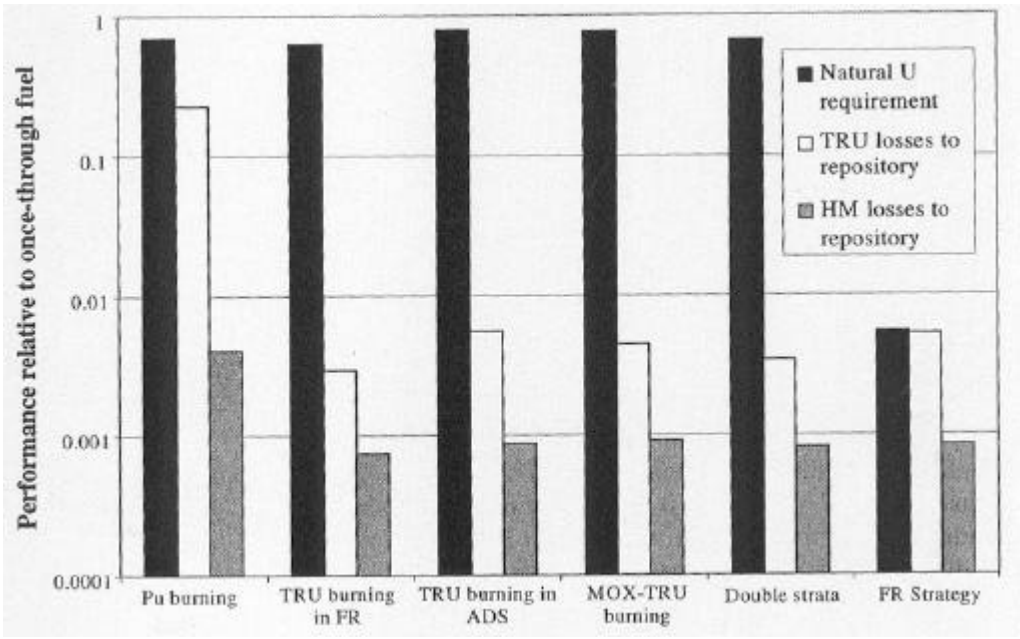

Figure 5.3. Resource efficiency and HLW production relative to open fuel cycle

The AAA study compared 9 options described in Figure 5.5, including deep burn gas cooled reactors, dual recycle MOX LWR's, TRU recycle MOX LWR's, fast reactors and ADS's. Note that the reactor designs were not optimized in all cases, and safety constraints were not necessarily respected. As was the case for the OECD study, neutronics methods were applied consistently, and generally accepted technological assumptions were used.

Main results are given below:

- Figure 5.6 provides the long term toxicity of the waste products of the various schemes. The behaviors in all cases are roughly similar, and later studies have shown that differences are mostly due to the normalization used in the study (results were normalized to one unit of initial SNF mass, whereas the OECD study was normalized to one unit of electricity produced: the US schemes penalizes approaches with fertile fuels

which produce some transuranics and thus increase the overall losses, but also produce more electricity)

- Figure 5.7 shows the influence of changing one assumption, the loss fraction, on the residual toxicity, and demonstrates the preponderance of this parameter.

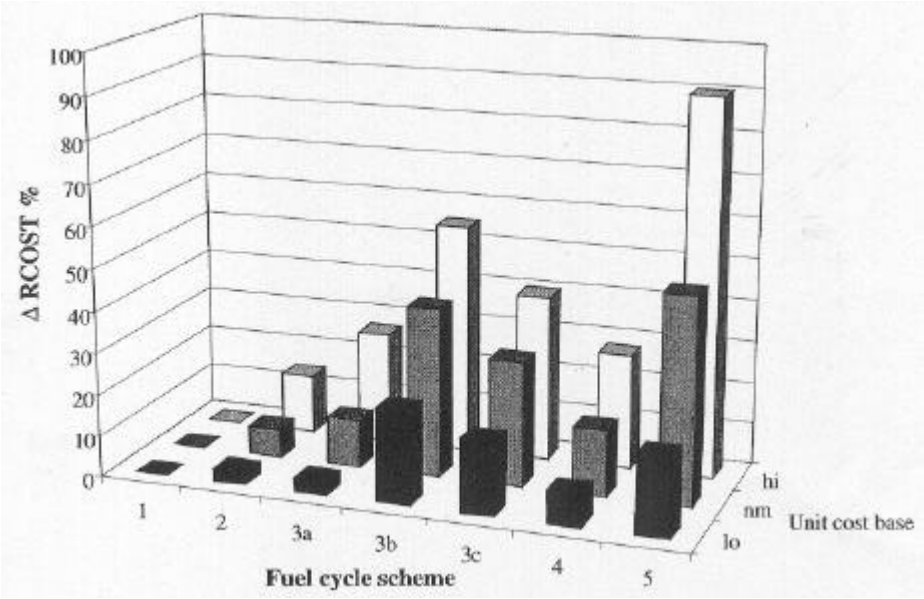

Figure 5.4. Impact of unit cost ranges for advanced-technology costs on the relative costs associated with each fuel cycle scheme

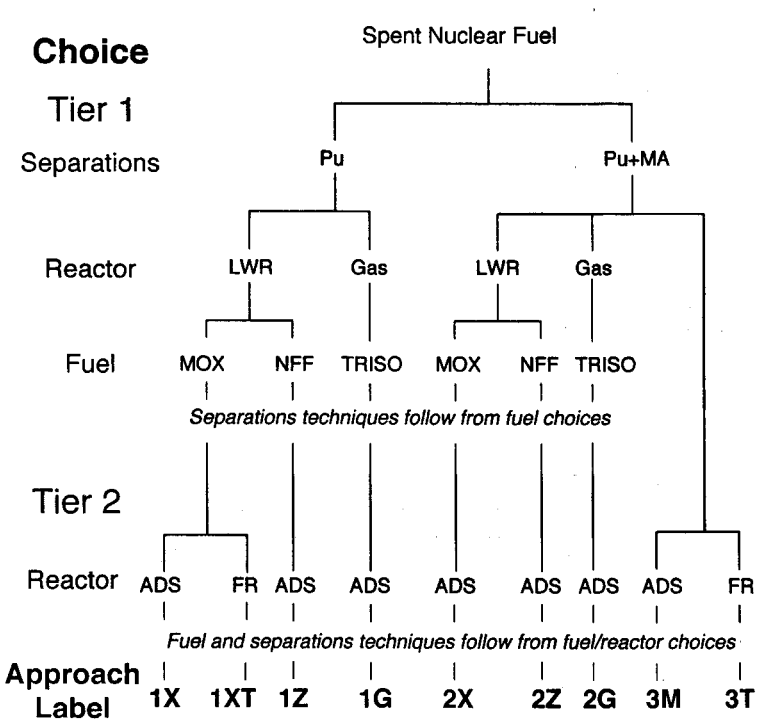

Figure 5.5. Approach tree

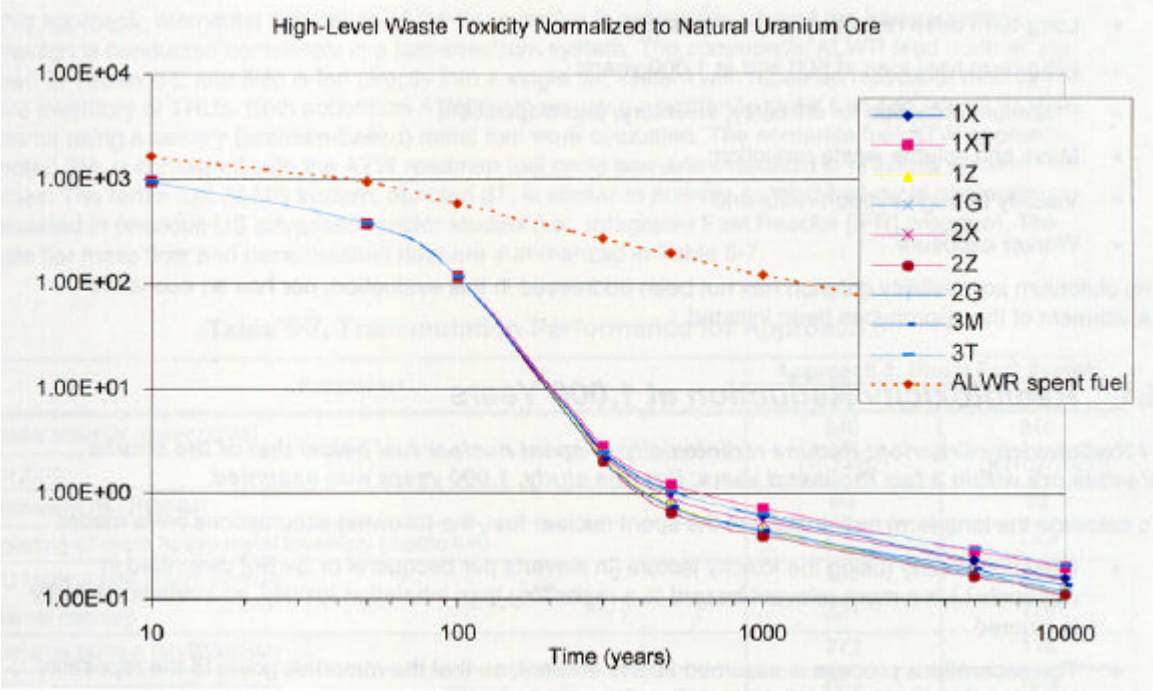

Figure 5.6. Comparison of long-term actinide toxicity of spent nuclear fuel and transmuted spent nuclear fuel to natural uranium ore

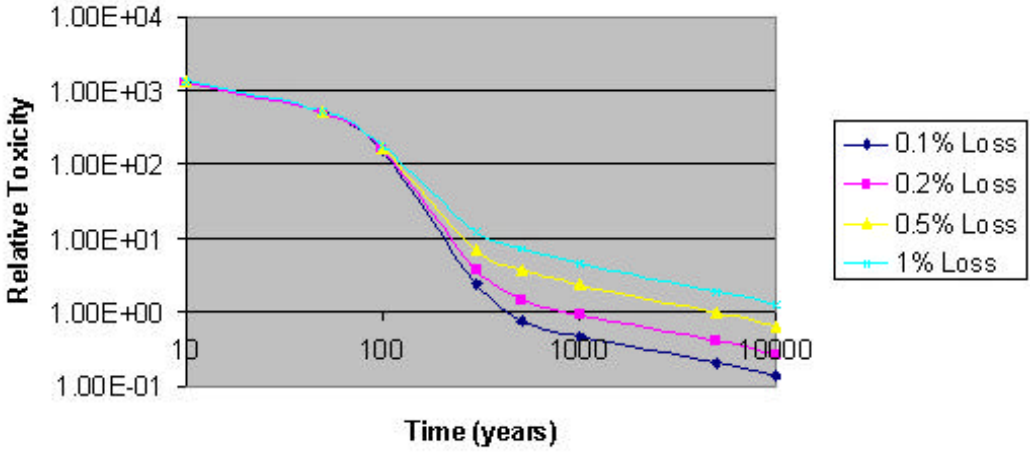

Figure 5.7. Toxicity

The general conclusions of these studies are the following:

1. All transmutation systems seem to have similar performances as far as final radiotoxicity is concerned;
2. The most important parameters for achieving strong toxicity reduction are the ability to achieve very low levels of losses in the fuels separations and reprocessing operations.
3. There appear to be significant differences in costs between the different scenarios, with a clear advantage for systems utilizing fast reactors rather than accelerator driven systems. Nevertheless, this result needs to be confirmed and put within the context of a national infrastructure strategy, which might favor the maximum use of existing commercial LWR's and a minimized second tier.

References

5.1 "Accelerator Driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles: a Comparative Study," published by the OECD Nuclear Energy Agency (2002).

5.2 G. Van Tuyle et al, "Scoping Evaluations of Candidate Approaches for an Integrated Waste Management Strategy," Trans. ANS, Vol. 86, pp 420-421 (June 2002).

Acknowledgements

Several colleagues provided material for this write up: Alan Baxter (General Atomics), Yousry Gohar (Argonne National Laboratory), Bob Hill (Argonne), John Stillman (Argonne), Mike Todosow (Brookhaven National Laboratory). Many thanks to them and their co-workers.

I made extensive use of knowledge acquired over the years from various organizations, in particular CEA and OECD, and would like to acknowledge their massive contributions to the progress of nuclear science.